

This electronic thesis or dissertation has been downloaded from the King's Research Portal at <https://kclpure.kcl.ac.uk/portal/>



Stochastic Representations of Open Systems

McCaul, Gerard Martin Gary

Awarding institution:
King's College London

The copyright of this thesis rests with the author and no quotation from it or information derived from it may be published without proper acknowledgement.

END USER LICENCE AGREEMENT



Unless another licence is stated on the immediately following page this work is licensed

under a Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International

licence. <https://creativecommons.org/licenses/by-nc-nd/4.0/>

You are free to copy, distribute and transmit the work

Under the following conditions:

- Attribution: You must attribute the work in the manner specified by the author (but not in any way that suggests that they endorse you or your use of the work).
- Non Commercial: You may not use this work for commercial purposes.
- No Derivative Works - You may not alter, transform, or build upon this work.

Any of these conditions can be waived if you receive permission from the author. Your fair dealings and other rights are in no way affected by the above.

Take down policy

If you believe that this document breaches copyright please contact librarypure@kcl.ac.uk providing details, and we will remove access to the work immediately and investigate your claim.

Stochastic Representations of Open Systems



Gerard McCaul

King's College London

A thesis submitted for the degree of

Doctor of Philosophy

2018

To anyone who can be bothered to read this.

Acknowledgements

First and foremost, I have to thank my supervisors Lev Kantorovich and Chris Lorenz. I could not ask for better guides, and would have failed on every level without their help, and occasional kicking. I'm sure reading the first draft of anything I've written is a real pain, so I owe them a debt of gratitude for that too. In that vein, I'd also like to thank Denys Bondar, first for agreeing to host my research placement, and second for offering me a postdoc despite the evidence of experience.

It would be impossible not to include my wife Sarah here, without whom I would probably be drinking myself to death in Shanghai. Thanks to her, I am drinking myself to death in London. I won't waste adjectives praising her, when it's sufficient to observe that no one else can understand why she hasn't dumped me yet. Also, sorry for writing this thesis while you were planning the wedding, it wasn't deliberate¹. I'd also like to express my gratitude to my parents for their continuing support and encouragement, and apologise for being such an awful teenager. *Plus ça change...*

Moving swiftly on, I'll always be grateful to Joe Bhaseen, Jemma Trick and Luke Simpson for being too weak-willed to say no to another drink. Guy Skinner deserves a mention for the many conversations on both the principles and practice of physics, and in his own words, "putting up with you for a week at APS". The last four years would undoubtedly have been diminished without the company of my CANES cohort. There are too many of you to list here, but I will say that before joining the centre I had no idea that Italy's main exports are olive oil and theoretical physicists. I'd also like to convey thanks to my fellow travelers in mid-twenties misery - Sam, Josh, Perky, and Bech. Without them, I would have been much more productive. Finally, in order to settle some petty scores, here are the organisations and people to whom I owe *nothing*, and wish to condemn in writing:

- The New Jersey state legislature for its puritanical licensing laws.
- Literally anyone who helped Jamie Oliver ban turkey twizzlers.
- The Roman Catholic church.
- The methhead I had to pay not to have dinner with me.

¹It was.

Abstract

This thesis outlines the development and implementation of an exact technique for the analysis of a particular class of open quantum systems. Starting from a generalised Caldeira-Leggett model, a set of coupled stochastic differential equations are derived as an evolution equation for the reduced density matrix of an arbitrary open system interacting (in a generalised manner) with a bath of harmonic oscillators. These equations are applicable even in the case of external driving and strong environment coupling. They also permit a more general class of initial states, where the combined system and environment are in full thermal equilibrium. Collectively these equations are known as the Extended Stochastic Liouville Equation (ESLE).

The ESLE is derived by casting the system+environment density matrix as a path integral in both real and imaginary time. In this form, it is possible to obtain the reduced system density matrix using influence functional techniques. Applying the two-time Hubbard Stratonovich transformation to this path integral, one obtains the ESLE. This consists of two evolution equations, accounting for a propagation in imaginary time followed by real time. Both equations contain stochastic terms which are non-trivially correlated and when averaged over realisations, give the exact reduced density matrix of the system.

A first application of the ESLE to a spin-boson model is also discussed. This is used as a proof of principle that the noises required by the ESLE can be generated numerically, and amenable to practical calculation. The impact of the ESLE's generalisations in the description of a two-level system being driven from equilibrium is also discussed.

An equivalent classical analysis is performed using Koopman-von Neumann (KvN) mechanics (an operational Hilbert space formalism which puts the quantum and classical descriptions on the same footing). In this setting, the ESLE derivation reproduces the Langevin equation directly from classical mechanics. Finally, the KvN formalism is used to explore some adjacent topics. In particular, a theory of classical self-adjoint extensions as a measure of local entropy conservation is developed.

Publications

- *Partition-free Approach To Open Quantum Systems In Harmonic Environments: An Exact Stochastic Liouville Equation*, G. M. G. McCaul, C. D. Lorenz, and L. Kantorovich, Phys. Rev. B **95**, 125124 (2017) - In this paper we derive the Extended Stochastic Liouville Equation (ESLE). This is a set of stochastic differential equations derived from a generalised Caldeira-Leggett model.
- *Driving Spin-boson Models From Equilibrium Using Exact Quantum Dynamics*, G. M. G. McCaul, C. D. Lorenz, and L. Kantorovich, Phys. Rev. B **97**, 224310 (2018) - We detail the numerical generation of noises with correlations across time dimensions for use with the ESLE, while the effect of a more general initial condition for a driven spin-boson model is also examined.
- *Entropy Non-Conservation for Hamiltonian Dynamical Systems*, Gerard McCaul, Alexander Pechen, and Denys I. Bondar (In Production) - This paper categorises classical entropy conserving distributions via KvN dynamics and the theory of self-adjoint extensions. As a result, we are able to produce several counter-examples of classical systems with non-conserved entropy evolving under Hamiltonian dynamics.
- *Classical Influence Functionals*, Gerard McCaul (In Production)- The KvN formalism is used to derive a classical path integral, from which a classical influence functional is constructed. Using this technique, a generalised Langevin equation is also obtained.

Contents

| | | |
|----------|------------------------------------------------------------------------|-----------|
| 1 | Introduction | 1 |
| 1.1 | Context | 1 |
| 1.2 | Fundamentals | 4 |
| 1.2.1 | Composite Systems | 5 |
| 1.2.2 | Time Evolution | 6 |
| 1.3 | Quantum Master Equations | 7 |
| 1.4 | Caldeira-Leggett Model | 12 |
| 1.5 | Thesis Outline | 15 |
| 2 | Methods | 18 |
| 2.1 | Path Integrals | 18 |
| 2.1.1 | Trotter Splitting | 20 |
| 2.1.2 | Expressing the propagator as a path integral | 24 |
| 2.1.3 | Time-Dependent Propagator | 26 |
| 2.1.4 | Semi-Classical Approximation | 29 |
| 2.1.5 | Propagator For The Free Particle | 31 |
| 2.1.6 | The Harmonic Oscillator | 32 |
| 2.1.7 | The Density Matrix as a Path Integral | 41 |
| 2.1.8 | The Influence Functional | 43 |
| 2.2 | Hubbard-Stratonovich Transformation | 47 |
| 2.2.1 | Real Gaussian processes | 47 |
| 2.2.2 | Complex Gaussian Distributions | 49 |
| 2.2.3 | The Multivariate Complex Gaussian | 52 |
| 2.2.4 | The Hubbard-Stratonovich Transformation | 55 |
| 2.2.5 | Hubbard-Stratonovich transformation over P time dimensions | 57 |
| 2.3 | Koopman von-Neumann Dynamics | 59 |
| 2.3.1 | The Koopman Operator | 59 |
| 2.3.2 | Basis overlaps | 62 |
| 2.3.3 | Liouville's Theorem For KvN | 64 |
| 2.4 | Chapter Summary | 69 |

| | | |
|----------|------------------------------------------------------------------|------------|
| 3 | The Extended Stochastic Liouville Equation | 70 |
| 3.1 | Model | 71 |
| 3.1.1 | Normal Modes | 73 |
| 3.2 | Representation with Influence Functional | 75 |
| 3.2.1 | Evaluating The Environmental Path Integral | 76 |
| 3.2.2 | Integrating Over Environment Terminals | 78 |
| 3.2.3 | The Influence Phase | 80 |
| 3.2.4 | Returning To The Real Space Representation | 82 |
| 3.3 | The Two-Time Hubbard-Stratonovich Transformation | 85 |
| 3.4 | The Extended Stochastic Liouville-von Neumann Equation | 88 |
| 3.4.1 | Effective Propagators | 90 |
| 3.4.2 | Real time Evolution | 91 |
| 3.4.3 | Imaginary Time Evolution | 92 |
| 3.4.4 | The ESLE | 97 |
| 3.5 | Derivations Using the ESLE | 98 |
| 3.5.1 | A Simplified Model | 99 |
| 3.5.2 | Classical Limit | 101 |
| 3.5.3 | Hamiltonian Of Mean Force | 106 |
| 3.5.4 | Attaching A Second Bath | 109 |
| 3.6 | Chapter Summary | 111 |
| 4 | Numerical Implementation | 113 |
| 4.1 | Generating Noises | 114 |
| 4.1.1 | Numerical Implementation Algorithm | 119 |
| 4.2 | spin-boson Models | 123 |
| 4.2.1 | Model Hamiltonian | 123 |
| 4.2.2 | Spin-boson Discretisation | 126 |
| 4.2.3 | Equilibrium Test | 126 |
| 4.3 | Landau-Zener Sweep | 129 |
| 4.3.1 | Temperature Dependence | 131 |
| 4.3.2 | Coupling-Strength Dependence | 133 |
| 4.3.3 | Comparison to Partitioned Evolution | 135 |
| 4.4 | Chapter Summary | 138 |
| 5 | Classical Analysis | 142 |
| 5.1 | The KvN path integral | 143 |
| 5.2 | Classical Influence Functional | 147 |
| 5.2.1 | Classical Model | 148 |
| 5.2.2 | Choice in the Hubbard-Stratonovich transformation | 153 |
| 5.3 | Alternative ESLE classical limit | 154 |
| 5.4 | Classical Self-Adjoint Extensions and Entropy | 159 |

| | | |
|----------|-------------------------------------------------------------------|------------|
| 5.4.1 | Self-Adjoint Operators | 161 |
| 5.4.2 | Entropy Conservation Using The Self-Adjoint Property | 163 |
| 5.4.3 | von-Neumann Deficiency index theorem | 164 |
| 5.4.4 | An Example: Simple Periodic system | 166 |
| 5.4.5 | Non-conserving Distribution In A Restricted Phase Space | 171 |
| 5.5 | Chapter Summary | 176 |
| 6 | Conclusions | 179 |

List of Figures

| | | |
|-----|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----|
| 1.1 | The evolution of a reduced density matrix $\hat{\rho}_Q$ can be described either as the trace over a unitarily evolved system, or equivalently as a dynamic map $\hat{\Lambda}$ | 7 |
| 1.2 | Schematic of a composite system, where the reduced dynamics of the Q system may be described with a dynamical map $\hat{\Lambda}$ | 9 |
| 2.1 | One dimensional evolution, where propagation is continuous in the left panel, and sliced into discrete sections on the right panel. | 21 |
| 2.2 | Geometric representation of limit swapping with hatching indicating the direction of the first integration | 39 |
| 3.1 | Example trajectory for the path integral in Eq. (3.16). Expressing the canonical density matrix as a path integral introduces a second time dimension, with dummy end-points \bar{q}, \bar{q}' | 75 |
| 3.2 | Representative trajectories for the evolution of the system. First there is an evolution in imaginary time up to $\tau = \beta\hbar$, before evolving in real time from this point up to time t_f . Different colours correspond to different simulations associated with particular manifestations of the noises. The average of the final points gives the physical density matrix at that time (indicated at time t_f). | 98 |
| 3.3 | Schematic demonstrating the full ensemble over different initial system+bath states. The braces indicate the result when averaging over the appropriate dimension. Averaging over both the ensemble and stochastic trajectory would yield the experimentally observed expectation $\langle \hat{A}(t) \rangle = A(t)$ | 105 |
| 4.1 | Typical correlation functions obtained from generated noise after 4×10^5 runs with parameters $\beta = 0.1\Delta^{-1}$, $\alpha = 0.2$, $\omega_c = 25$ and an Ohmic spectral density. | 122 |

- 4.2 ESLE evolution for a fast sweep with $t_0 = -6\Delta^{-1}$, $\kappa = 4\Delta^2$, and $\alpha = 0.05$, sampled over 3×10^5 runs using 2^{14} and 2^{16} steps. The larger timestep (the green curve) leads to an unphysical magnification in the amplitude of oscillations (although the mean values are still comparable). 127
- 4.3 ESLE evolution of ρ for a time-independent system, averaged over 1×10^5 runs (solid lines). Dashed lines indicate an equivalent SLE evolution from $\rho_{ij} = \delta_{i1}\delta_{j1}$, while dash-dotted lines (not seen as they coincide with the solid lines) are SLE evolutions from $\rho_{ij} = \langle \bar{\rho}_{ij}(\hbar\beta) \rangle_r$. Here $\Delta = 1$, $\epsilon = 5\Delta$, $\beta = 0.1\Delta^{-1}$, $\alpha = 0.05$, and $\omega_c = 200$. While the ESLE shows small fluctuations, the density matrix remains on average constant. The SLE evolution from $\rho_{ij} = \delta_{i1}\delta_{j1}$ predicts a relaxation to the ESLE values, but on a timescale not accessible by the simulation. 128
- 4.4 ESLE evolution of $\langle \sigma_z \rangle$ for a fast sweep with $t_0 = -6\Delta^{-1}$, $\kappa = 4\Delta^2$, and $\alpha = 0.05$, sampled over 3×10^5 runs, calculated at three values of the temperature. Red and blue dashed lines indicate extrapolated asymptotes for the two lowest temperature simulations. We observe that even at weak coupling the asymptotic value of $\langle \sigma_z \rangle$ deviates from the LZ expectation $\langle \sigma_z \rangle_{LZ}$, although lower temperature asymptotic states lie closer to the LZ limit. 133
- 4.5 (a) Real time $\langle \sigma_z \rangle$ dynamics for the system with parameters $t_0 = -6\Delta^{-1}$, $\kappa = 5\Delta^2$, and $\beta = 0.2\Delta^{-1}$, after sampling with 5×10^5 runs. (b) The same dynamics are rescaled such that the curves of different α lie nearly on top of one another, demonstrating the spin-bath coupling renormalising the tunneling element of the two level system. Higher coupling strengths require more averaging to reach longer times, and hence have been truncated. 134
- 4.6 Real time spin dynamics for the system with parameters $t_0 = -6\Delta^{-1}$, $\kappa = 5\Delta$, $\beta = 0.2\Delta^{-1}$, sampled over 5×10^5 runs. (a) and (b) show $\langle \sigma_z \rangle$ at coupling strengths $\alpha = 0.05$, and $\alpha = 0.2$. respectively. 135
- 4.7 Real time spin dynamics of $\langle \sigma_x \rangle$ for the system with parameters $t_0 = -6\Delta^{-1}$, $\kappa = 5\Delta$, $\beta = 0.2\Delta^{-1}$, $\alpha = 0.05$ sampled over 5×10^5 runs. Except for the initial oscillations, the behaviour in all simulations appears identical. 136
- 4.8 The evolution of the difference in $\langle \sigma_z \rangle$, calculated relative to the ESLE result of Fig. 4.6, at a) $\alpha = 0.05$, and b) $\alpha = 0.2$. The dashed lines indicate the mean value of the differences calculated using the final eighth of the simulation. 137

| | | |
|-----|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----|
| 4.9 | Distribution of σ_z at $t_0 = -6\Delta^{-1}$, $\kappa = 5\Delta^2$, $\beta = 0.2$ and $\alpha = 0.05$, sampled over 5×10^5 runs. As time progresses, the noise envelope drastically increases the width of the distribution, which requires more sampling to accurately calculate its mean. | 138 |
| 5.1 | An operator A and its conjugate have different domains. A self adjoint extension modifies the domains of both operators such that they are identical. | 166 |
| 5.2 | Example phase-space boundaries in (left) original coordinate system and (right) action-angle coordinates. | 167 |
| 5.3 | In the harmonic oscillator, time evolution is equivalent to rotation. Here the red curve represents the transport of $\psi(\theta_-, J, t)$ in time for a constant J . The blue arrows denote the phase due to the self-adjoint extension, where after each period an additional phase of $e^{i\beta(J)}$ is picked up. | 170 |
| 5.4 | Normalisation time-dependence $\theta_+ - \theta_- = \theta_b$ | 174 |
| 5.5 | Entropy rate of change. | 174 |
| 5.6 | In this case we see $\langle J(t) \rangle$ is also a non-conserved quantity. | 175 |
| 5.7 | Distribution evolving with time. We can consider this as either a fixed excluded region with a rotating distribution, or a fixed distribution with an evolving excluded region. The red curve is a line of constant J , and the difference between the two boundary points on these curves is the essential contribution to entropy non-conservation. | 176 |

Chapter 1

Introduction

As an adolescent I aspired to lasting fame, I craved factual certainty, and I thirsted for a meaningful vision of human life- so I became a scientist. This is like becoming an archbishop so you can meet girls.

Matt Cartmill

1.1 Context

The predictive power of physics rests on the presumption of universal laws. These include global spatial and temporal symmetries which demand momentum and energy conservation [1], while time reversal symmetry arises as a consequence of Hamiltonian dynamics [2]. Problematically however, we do not see the conservation implied by fundamental symmetries in mundane experience. Energy leaks, structure deteriorates, and lifetimes (both correlative and biological) are finite. This is an altogether antique notion - “all human things are subject to decay/And when fate summons, monarchs must obey” [3] - but time has not diminished its essential truth.

Physical systems often display the same characteristics of dissipation and fluctuation familiar to us in everyday life, and a great many quantum phenomena cannot be explained without reference to them [4–6]. These behaviours are the consequence of the impossibility of isolating any realistic system from an uncontrolled environment. Collectively, such systems are termed *open systems*, and they constitute an essential part of both classical and quantum theory.

Embedding a consistent theory of open systems within the quantum regime has not historically been a straightforward process however. Early attempts followed the usual procedure of taking an effective Hamiltonian displaying the requisite dissipative properties and quantising it. A typical example of this is the early work of Kerner and Stevens on sets of damped harmonic oscillators [7,8]. The basis of this method in classical phenomenological equations means that it is capable of providing exact solutions for some simple systems, such as the damped harmonic oscillator. These solutions are however undermined by being intrinsically incompatible with quantum mechanics. This arises from the fact that there are no *time-independent* Hamiltonians that can replicate the equation of motion for a damped oscillator,

$$m\ddot{x} + \alpha\dot{x} + m\omega^2x = 0 \tag{1.1}$$

which has frequency ω and friction α . While there exists a *time-dependent* Hamiltonian that leads to this equation of motion [9], after quantisation the fundamental commutation relation has a decaying time-dependent form [10]. This unphysical result illustrates some of the difficulties in tackling the description of open quantum systems. Unlike the classical case, any phenomenological approach must *also* obey the non-trivial constraints of a non-commutative algebra.

Naturally, this set-back did not mark the end of the study of open quantum systems, and the following decades have brought to us a broad corpus of techniques and insights for their analysis. This process is ongoing, and of some importance to

both fundamental and applied research. Phenomena fundamental to our understanding of the quantum-classical transition (such as decoherence) cannot be observed or explicated without recourse to environments [11]. They are an essential component of quantum thermodynamics [12, 13], and vital to the field of quantum computing, where coherence is a resource which the environment can dissipate [14–16] or *enhance* with suitable environmental engineering [17]. Technology is rapidly reaching a scale where the effect of quantum environments must be accounted for, and developing sufficiently powerful descriptions of them is necessary to fulfill the promise of a wide array of disciplines.

The goal of this thesis is to expound a particular methodology describing the evolution of open systems under more general conditions than previously derived. Naturally, it is necessary to set these results in their proper context. There are many excellent reviews on the theory of open quantum systems [4, 18–20], but sections 1.2–1.4 attempt to distil the essential detail necessary to understand the rest of the thesis, with particular emphasis placed on work similar (in its goals or methodology) to our own. Section 1.5 will then outline the structure of latter chapters.

Ultimately much of this document will consist of mathematics, and lengthy derivations do not lend themselves to elegant prose, except possibly as a test of one’s lexical reserve of conjunctions¹. Periodically however, there are results which touch on broader themes, and lend themselves a little better to digression. As such I have shamelessly seized these opportunities to extemporise, but won’t pretend my personal collection of prejudices is essential reading. As such I have marked these passages as *asides*, which the reader not partial to sybaritic diatribes may safely ignore.

Finally, given the preponderance of equations, the most important results have been framed in the following manner to distinguish them:

¹Pity the Choctaw scientist, whose language lacks conjunction as a separate operation from subordination. It does however possess a large class of subordinators that do the work of conjunctions [21].

1.2 Fundamentals

The fundamental object of interest for open quantum systems is the *density matrix*, defined as

$$\hat{\rho} = \sum_{\alpha} p_{\alpha} |\psi_{\alpha}\rangle \langle \psi_{\alpha}|. \quad (1.2)$$

The density matrix is a statistical mixture of pure states ψ_{α} , and generalises the notion of a quantum state to systems also governed by classical probability distributions. Typically, this is necessitated by the need to describe a thermal system, where the energy eigenstates are weighted by the Gibbs distribution. Thus, a common density matrix is the canonical/thermal/Gibbs² density matrix:

$$\hat{\rho}_{\beta} = \frac{1}{Z_{\beta}} \sum_{\alpha} e^{-\beta E_{\alpha}} |E_{\alpha}\rangle \langle E_{\alpha}| = \frac{1}{Z_{\beta}} e^{-\beta \hat{H}} \quad (1.3)$$

$$Z_{\beta} = \sum_{\alpha} e^{-\beta E_{\alpha}}. \quad (1.4)$$

The relationship of a density matrix to an expectation is easily verified,

$$\langle \hat{A} \rangle = \text{Tr} [\hat{\rho} \hat{A}] = \int dx A(x) \rho(x, x) \quad (1.5)$$

where the final equality expresses the density matrix in a specific basis:

$$\rho(x, x') = \sum_{\alpha} p_{\alpha} \langle x' | \psi_{\alpha} \rangle \langle \psi_{\alpha} | x \rangle. \quad (1.6)$$

²This object has acquired a large collection of aliases due to its ubiquity across physics, and I will use them interchangeably.

Note that in the case of \hat{A} being the unit operator, we obtain $\text{Tr}[\hat{\rho}] = 1$.

There are a great many revealing properties of quantum systems which may be derived purely from the density matrix. The non-existence of a dispersion-free (classical) ensemble described by the density matrix follows almost directly from its definition [22], and its ability to treat composite systems provides a useful measure of entanglement [18]. While neither of the aforementioned properties are directly relevant to the work we shall present, the density matrix approach to composite systems is an essential element to modelling open systems.

1.2.1 Composite Systems

Consider two interacting quantum systems Q and X . The individual systems are described with Hilbert spaces of dimension \mathcal{H}_Q and \mathcal{H}_X respectively. The Hilbert space of the combined system will be the tensor product of the individual systems, $\mathcal{H} = \mathcal{H}_Q \otimes \mathcal{H}_X$, and the density matrix will be denoted by $\hat{\rho}_{\text{tot}}$. If one is interested only in the observables of one of the subsystems (say Q), then in the combined space it is given by $\hat{A}_Q \otimes \hat{I}_X$ where \hat{I}_X is the unit operator of the appropriate dimension. The expectation of this is

$$\langle \hat{A}_Q \rangle = \text{Tr} [\hat{A}_Q \otimes \hat{I}_X \hat{\rho}_{\text{tot}}] = \text{Tr}_Q [\hat{A}_Q \text{Tr}_X [\hat{\rho}_{\text{tot}}]] \quad (1.7)$$

where in the final equality $\text{Tr}_{Q/X}$ refers to a trace over the appropriate subspace. This equation for subsystem expectations also means the density matrix describing that subsystem is uniquely defined:

$$\hat{\rho}_Q = \text{Tr}_X [\hat{\rho}_{\text{tot}}]. \quad (1.8)$$

This is the *reduced density matrix*. A composite system serves as a model for an interacting open system and environment, but it is only the dynamics of the open

system that we are interested in³. The reduced density matrix is therefore the key descriptor of open quantum systems.

1.2.2 Time Evolution

Quantum states are evolved by a *propagator* \hat{U}

$$|\psi(t)\rangle = \hat{U}(t) |\psi(0)\rangle \quad (1.9)$$

whose general form is

$$\hat{U}(t) = \hat{T} \exp \left[-\frac{i}{\hbar} \int_0^t dt' \hat{H}(t') \right] \quad (1.10)$$

where \hat{T} is the time ordering operator and \hat{H} is the system Hamiltonian. In the case of a density matrix, its evolution is described by the *Liouville equation*

$$i\hbar \frac{d}{dt} \hat{\rho}(t) = [\hat{H}, \hat{\rho}] \quad (1.11)$$

with the formal solution

$$\hat{\rho}(t) = \hat{U}(t) \hat{\rho}(0) \hat{U}^\dagger(t). \quad (1.12)$$

An important feature of time evolution is that it is a *unitary* transformation, which excludes precisely the phenomena that characterise open systems. The dynamics of a subsystem of the density matrix has a more complex form as it is described by the reduced density after time evolution

$$\hat{\rho}_Q(t) = \text{Tr}_X \left[\hat{U}(t) \hat{\rho}(0) \hat{U}^\dagger(t) \right]. \quad (1.13)$$

³For the simple expedient that it is the often the only part of the joint system that we can effectively control.

The partial trace over a system is *not* a unitary operation, and opens the door to dissipative, fluctuating behaviour in the reduced system. In principle, Eq. (1.13) is a complete description of the dynamics of any open system, but is entirely unilluminating. In large part the study of open quantum systems is simply re-expressing this equation in a more useful form.

One of the principal approaches to the problem of describing the evolution of a reduced density matrix is to use a *dynamical map* $\hat{\Lambda}(t)$:

$$\hat{\rho}_Q(t) = \hat{\Lambda}(t) \hat{\rho}_Q(0) \quad (1.14)$$

This map has fewer constraints on it than a unitary operator, with the only requirement being that it must be *completely positive* and *trace preserving* [23]. Figure 1.1 shows the relationship between the two forms of evolving the reduced density matrix.

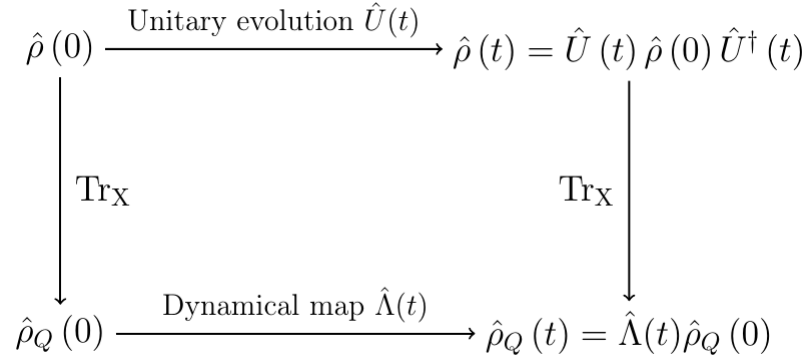


Figure 1.1: The evolution of a reduced density matrix $\hat{\rho}_Q$ can be described either as the trace over a unitarily evolved system, or equivalently as a dynamic map $\hat{\Lambda}$.

1.3 Quantum Master Equations

Elucidating the dynamics of the reduced system is often the ultimate objective for any system we study. Very often, dissipative systems are modelled as a primary

system (the “open system”, Q) of interest coupled to an explicit secondary system (the “environment” or “heat bath”, X) which together describe the overall system being modeled (the “total system”). This approach was pioneered by Callen, Welton, Senitzky and Lax [10, 24, 25], using a microscopic Hamiltonian to describe this:

$$\hat{H}_{\text{tot}} = \hat{H}_Q \otimes \hat{I}_X + \hat{I}_Q \otimes \hat{H}_X + \hat{H}_I \quad (1.15)$$

where \hat{H}_I is the interaction between the two systems. A schematic of this joint system can be seen in Fig. 1.2. Since the combined system evolves unitarily, it obeys the standard rules of quantum mechanics. The dynamics of the reduced system will be described by a dynamical map, incorporating both the dissipation *and* fluctuations experienced by the open system as a consequence of its explicit coupling to the environment. Combining this model with appropriate system specifications allows differential equations to be derived which retain the correct behaviour in the classical limit [26–30]. These equations are referred to as *quantum master equations*, in analogy with their classical counterpart.

Perhaps the most famous quantum master equation is the *Lindblad equation*. This does not describe a specific system, but is instead the most general possible master equation under certain simplifying assumptions [18]. Chief among these is the assumption that the family of dynamical maps forms a *semigroup*:

$$\forall t_1, t_2 > 0, \quad \hat{\Lambda}(t_1) \hat{\Lambda}(t_2) = \hat{\Lambda}(t_1 + t_2). \quad (1.16)$$

Physically, this corresponds to demanding that the system is *Markovian*. Generally speaking, this presumption of a memoryless evolution of the reduced system is justified if the environment correlations decay on a much shorter timescale than the open system’s dynamics. If the dynamical maps are also continuous, then it is possible to

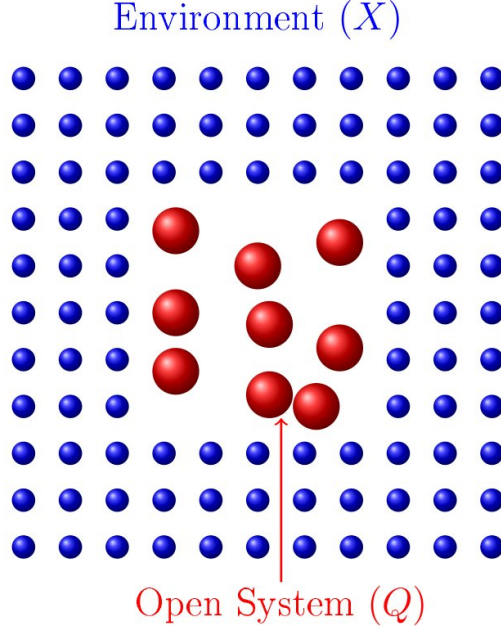


Figure 1.2: Schematic of a composite system, where the reduced dynamics of the Q system may be described with a dynamical map $\hat{\Lambda}$.

express the time evolution of the reduced system with the generator \mathcal{L} of the map:

$$\hat{\Lambda}(t) = e^{\mathcal{L}t} \quad (1.17)$$

$$\frac{d\hat{\rho}_Q}{dt} = \mathcal{L}\hat{\rho}_Q. \quad (1.18)$$

With these assumptions, the Lindblad equation gives the most general form for the evolution of $\hat{\rho}$ [31]:

$$\frac{d\hat{\rho}_Q}{dt} = -i \left[\hat{H}, \hat{\rho}_Q \right] + \sum_k \gamma_k \left(\hat{A}_k \hat{\rho} \hat{A}_k^\dagger - \frac{1}{2} \left\{ \hat{A}_k^\dagger \hat{A}_k, \hat{\rho}_Q \right\} \right). \quad (1.19)$$

There are a great many remarks that might be made on the Lindblad equation, as it has been subjected to extensive analysis both mathematical and physical [32]. The most important point is that the Lindblad equation is the result of an existence theorem, and offers no interpretation for its components. The first term in the equation

is a unitary evolution, so it is natural to associate \hat{H} with some system Hamiltonian. The second term is more difficult to interpret physically, but the γ_k can be considered decay rates for the different channels of relaxation available to the system [18]. The operators \hat{A}_k are known as the Lindblad (or jump) operators. Their exact physical meaning is unclear, but they are associated with the fluctuations one expects in an open system. It is always possible to interpret the Lindblad form as a system undergoing continuous measurement [33], enabling a simple derivation of the quantum Zeno effect [34]. Another significant feature of the Lindblad equation is that it furnishes quantum theory with an H theorem [35]. This states that the increase in the von-Neumann entropy $S = -\text{Tr}[\hat{\rho}\ln(\hat{\rho})]$ for a density matrix evolving under the Lindblad equation is always greater than zero. The Lindblad equation therefore plays an important role in generalising thermodynamic concepts to quantum systems and establishing an analog to the second law.

The Lindblad equation is far from the only way to interrogate the dynamics of an open system. Even if we do wish to represent dynamics in a Lindblad form, a microscopic model is necessary to identify not only the decay rates and Lindblad operators, but also the \hat{H} operator (it is not necessarily the case that the Hamiltonian in Eq.(1.19) is the free Hamiltonian \hat{H}_Q of the reduced system). For a composite system of the type described by Eq.(1.15), the progress one is able to make is constrained by the explicit form of the environment and interaction. In general, finding and solving the master equation for the reduced system is no less difficult than simply analysing the full system-environment amalgam. It is therefore usually wise to adopt several simplifying assumptions.

We have already encountered the Markovian approximation, but in addition to this the environment coupling is often taken to be *weak*. This amounts to stipulating that the environment is time-independent and minimally affected by the interaction,

such that the total system density matrix may at any time be approximated by:

$$\hat{\rho}(t) \approx \hat{\rho}_Q(t) \otimes \hat{\rho}_X \quad (1.20)$$

This is known as the *Born approximation* and appears in concert with the Markovian assumption often enough that taken together they are labeled the *Born-Markov approximation*. With these simplifications, it is possible to derive (in the interaction picture) the *Redfield equation* [36,37]:

$$\frac{d}{dt}\hat{\rho}_Q(t) = - \int_0^\infty dt' \text{Tr}_X \left[\hat{H}_I(t), \left[\hat{H}_I(t-t') \hat{\rho}_Q(t) \otimes \hat{\rho}_X \right] \right]. \quad (1.21)$$

This formulation is particularly useful when combined with a secular approximation [18]. This is closely related to the rotating wave approximation (which neglects rapidly oscillating terms) [38], and as such the Redfield equation has been utilised extensively in studies of dissipative quantum optics [39].

Let us return for a moment to the assumption of Markovianity. Given so many master equations are predicated on it, we should expect it is a reasonable simplification. Unfortunately, Markovian behaviour has unexpected consequences for quantum thermodynamics. In the classical case the Langevin [40] and Fokker-Planck [41] models are based on a memoryless interaction and thermalise systems⁴. In the quantum case however, it has long been postulated that translationally invariant Markovian *quantum* systems do not thermalise [43]. This statement has been the cause of some controversy [44], particularly as the free Brownian particle evolving under a Lindblad equation *does* thermalise [45].

The free particle represents a singular case however, and recently this long suspected no-go theorem was proven in general [46]. The demonstration of non-thermalisation for *all* Markovian models is a profound issue, since any physically reasonable model

⁴There are a few exceptions to this statement [42].

must equilibrate with its environment in the long time limit. The fact that this is impossible for memoryless models suggests they do not exist in nature, and any thermodynamic information concluded from such models cannot be relied upon!

The master equations presented in this section are generic, but their generality comes at the cost of some rather severe (and intrinsically unphysical) approximations. In the next section we shall examine a particular microscopic model, which by virtue of its specificity does not require these restrictions.

1.4 Caldeira-Leggett Model

As we have seen, there are a number of general schemes to describe open systems. Ultimately however, one must parametrise their model in some way. This is often done by choosing a specific microscopic Hamiltonian to represent the system and environment. In practice the functional form of the environment (secondary system) and its coupling must be chosen subject to several conditions. We are for example constrained by the expectation that in the high-temperature limit we recover a classical Brownian motion. In addition, if the elimination of environmental coordinates is to be exact, yet analytically tractable, the choice of environment and coupling is extremely limited. It is these considerations that motivate the popular Caldeira-Leggett (CL) Hamiltonian [47]:

$$H = H_Q(q) + \frac{1}{2} \sum_n (m_n \dot{x}_n^2 + m_n \omega_n^2 x_n^2) - q \sum_n c_n x_n + \frac{q^2}{2} \sum_n \frac{c_n^2}{m_n \omega_n^2}. \quad (1.22)$$

This model couples the open system (described by the coordinate q) to an environment of independent harmonic oscillators (masses m_n , frequencies ω_n , and displacement coordinates x_n) with each oscillator being coupled to the open system with a strength

c_n . The final term is a counter-term included to enforce translational invariance on the system and eliminate quasi-static effects [48].

One of the key advantages of this model is that it is possible to do away with many of the assumptions required to formulate the system agnostic master equations discussed previously [11]. As a consequence of this, the master equation for the CL model *cannot* be brought to the Lindblad form in Eq.(1.19) [18].

A particularly successful line of attack for the CL model is with path integrals. Path integrals remove the need for an explicit quantisation of the system Hamiltonian, as in this formalism quantum-mechanical propagators are represented as phase-weighted sums over trajectories, where the phase associated to each trajectory is proportional to the action of that path in the classical system [49]. A useful consequence of this is that the classical limit is easily obtained [50], and the quantisation of the system is automatic when choosing this representation. Finally, and probably most importantly, bath degrees of freedom can be integrated out exactly if the environment is harmonic and interacts with the open system via an expression that is at most up to the second order in its displacements. Some specific successful applications include tunnelling and decay rate calculations (Kramer’s problem) [5,6,51–54] as well as recent first-principle derivations for the rate of processes in instanton theory [55,56].

The Feynman-Vernon influence functional formalism [57] has spawned a great many techniques based on the exact integration of the environment using path integrals. Much work has been done using this formalism, expanding the methodology of the Feynman-Vernon influence functional for both exact and approximate results [58–60].

Using the influence functional does not require the Born-Markov approximation, only the weaker condition that *initially* the density matrix of the total system $\hat{\rho}_0$ can be partitioned:

$$\hat{\rho}_0 = \hat{\rho}_Q(0) \otimes \hat{\rho}_X(0). \quad (1.23)$$

While this is not as extreme as the Born approximation, it still mandates that the open system and bath are initially non-interacting before being turned on for the dynamical evolution. This *partitioned* approach, strictly speaking, is still only applicable to weak system-bath coupling (at least during the transient regime of the evolution). This significantly limits the applicability of theories based on this approximation, but there are several approaches to circumventing this condition.

In the case of a two-state system, it is possible to approximate strong coupling directly using a reaction coordinate mapping [61]. This takes a system strongly coupled to an environment and maps it to an enlarged system, where the open system is now only coupled to a single reaction coordinate. This reaction coordinate is itself weakly coupled to the mapped environment, allowing the weak coupling approximation to be applied. This mapping has been successfully applied to models of qubits for quantum control [62] and heat engines at strong coupling [63].

More generically, in the CL model it is also possible to represent a strongly coupled initial state via an imaginary time path integral. This possibility was first noted by Smith and Caldeira [58], before being properly exploited by Grabert, Ingold and Schramm [64], who derived the time dependent expression for the reduced density matrix of an open system where all path integrals associated with the environment are fully eliminated.

With this corpus of techniques, path integrals (and specifically influence functionals) represent a powerful and flexible formalism that can be used to attack the problem of open quantum systems. Using the influence functional model, quantum Langevin equations for the reduced density matrix have been rigorously derived using path integrals [11, 47, 65–68]. In special cases, further analytical results have also been obtained by Kleinert [69, 70] and Tsusaka [71]. Generalisations of these results

to anharmonic baths produce approximate but more realistic models [72, 73], while time-dependent heat exchange can also be exactly included [74]. Approaches based on influence functionals have also found use in the real time numerical simulations of dissipative systems [75–81]. Alloying the influence functional with stochastic transformations has motivated other novel approaches, including the stochastic Schrödinger Equation [82, 83], quasiadiabatic path integral [84] and the Stochastic Liouville-von Neumann Equation (SLE) [85].

The SLE is particularly relevant in this context, as the generalisation of this technique is one of the principle results discussed in this manuscript. Briefly, when one combines the CL model influence functional with the *Hubbard-Stratonovich transformation* (see section 2.2) it is possible to exactly map the reduced system dynamics to the stochastic average of the following equation

$$i\hbar \frac{d\tilde{\rho}(t)}{dt} = \left[\hat{H}_Q(t), \tilde{\rho}(t) \right] - \eta(t) [\hat{q}(t), \tilde{\rho}(t)] + \gamma [\hat{q}^2(t), \tilde{\rho}(t)] + \frac{\hbar}{2} \nu(t) \{ \hat{q}(t), \tilde{\rho}(t) \}. \quad (1.24)$$

Here η and ν are complex Gaussian noises, while γ is a constant. Both the noise correlations and γ are determined entirely by the CL model parameters, and the average over noise realisation is $\langle \tilde{\rho}(t) \rangle = \hat{\rho}_Q(t)$. The SLE is a powerful, compact expression that precisely captures the effect of a harmonic environment, however it is not the most general formulation possible. The route to this generalisation (and therefore the structure of the thesis as a whole) will now be outlined.

1.5 Thesis Outline

We now motivate our own work in the context of open quantum systems. Recently, a more general version of the CL Hamiltonian (the open system and harmonic environment) was introduced [86] which is only linear with respect to the environmental

variables, but remains arbitrary with respect to the positions of atoms in the open system (this model is detailed in section 3.1). In this Hamiltonian interactions within the environment are not diagonalised. This is useful because all parameters of the environment and its interaction with the open system can then be extracted by expanding the Hamiltonian of a realistic combined system in atomic displacements of the bath and retaining only harmonic terms. This rather general choice of total system Hamiltonian enables one to derive classical equations of motion for the atoms in the open system [86] and propose an efficient numerical scheme for solving them [87–89].

This method has also been generalised to the fully quantum case [90] where it was shown, using a method based on directly solving the Liouville equation, that equations of motion for the observable positions of atoms in the open system have the form of a generalised Langevin equation with a friction kernel and non-Gaussian random force terms. Although this method enables one to develop the general structure of the equations to be expected for the open system, it lacks an exact mechanism for establishing the necessary expressions for the random force correlation functions.

Almost every method mentioned in this chapter is based on initially partitioning the total system. The initial condition of Eq. (1.23) is not always realistic however, as ordinarily one would be unable to “prepare” a quantum system with the interaction between the open system and environment switched off, prior to any perturbation being applied (one exception to this would be the initial open system and environment interaction Hamiltonians being identical - in this case the effect of the environment is simply a scaling of the open system Hamiltonian). As a result, the transient behaviour we predict for perturbations away from a partitioned initial condition will usually be spurious due to the artificial initial equilibration of each system separately. If we wish to extract the exact transient dynamics of an open system under general conditions we must use a more realistic, non-partitioned initial condition.

Fortunately the influence functional formalism has the capacity to accommodate

the generalisation of both the CL Hamiltonian and its initial conditions, rendering the assumption of a partitioned initial state unnecessary. It is therefore possible to derive an approximation-free description of any CL Hamiltonian. In chapter 2 the mathematical methods necessary to derive these new results are presented. Chapter 3 derives the Extended Stochastic Liouville Equation (ESLE), a generalisation of Eq. (1.24). This consists of a set of stochastic differential equations for the reduced density matrix of the open system, derived without requiring either the partitioned initial condition or weak bath coupling approximations. Several subsequent results using the ESLE are also discussed, including a heuristic classical limit. Chapter 4 presents a first application of the ESLE, modeling a driven two state system numerically. In chapter 5, a classical analysis is performed using the same influence functional techniques in a classical Hilbert space formalism. This results in both a rigorous classical limit for the ESLE, and a novel derivation of a generalised classical Langevin equation. The same formalism used to perform the classical analysis is then applied to the question of entropy conservation in classical systems. The thesis is closed with a discussion of the collected results in chapter 6.

Chapter 2

Methods

What has been will be again, what
has been done will be done again;
there is nothing new under the sun.

Ecclesiastes 1:9

This chapter outlines the mathematical techniques used to obtain the results of later chapters. The principal topics covered here are path integrals (including the influence functional), the HS transformation and Koopman-von Neumann dynamics. The first two sections are relied upon for the derivation covered in chapter 3, while chapter 5 employs the Koopman-von Neumann formalism.

2.1 Path Integrals

The path integral is an alternative representation for quantum mechanics, where the evolution of abstract states in a Hilbert space is replaced by a functional integral over classical trajectories. The spiritual predecessor of this approach is the Wiener integral, another functional integral designed to describe classical Brownian motion with a sum over stochastic trajectories [91]. In the quantum regime, the original inspiration for

the path integral can be traced to Dirac [92, 93].

The essential motivation for this was the observation that standard quantum mechanics is founded on an analog to classical Hamiltonian dynamics, but that classically this formulation is not unique. Specifically, by performing a Legendre transform on the Hamiltonian, one obtains the Lagrangian representation [2]. While these two approaches make equivalent predictions, the Lagrangian method possesses a few key advantages. First, system dynamics are more compactly described in the Lagrangian method via the principle of stationary action, which has no Hamiltonian equivalent. Additionally, the action of a system is a relativistically invariant quantity [94], whereas the Hamiltonian form carries a particular time coordinate as its canonical conjugate. This means Lagrangian formulation is equipped with the desirable property that it is easier to incorporate generalisations.

Generalising the Lagrangian approach to include quantum mechanics is far from trivial however, given that its equations of motion use partial derivatives of the Lagrangian with respect to both coordinates and velocity while quantum mechanics can only tolerate derivatives with respect to the canonical dynamical variables. Dirac's approach to this problem was to consider the overlap between two states at different times. One state is propagated forward in time, and its propagator may be Legendre transformed at each point in time. Following this prescription, one finds that the overlap is proportional to $e^{i\frac{S}{\hbar}}$, where S is the action $S = \int_0^{t_f} L[x(\tau), \dot{x}(\tau)] d\tau$. While this result captures the essential behaviour of the path integral, an explicit prescription for constructing it (and a proof of its consistency with both the Schrödinger equation and fundamental commutation relations) was not achieved until Feynman's seminal contributions [49].

This path integral possesses a few principal advantages over the standard quantum mechanical formalism. The aforementioned relativistic invariance, as well a transparent manifestation of the correspondence principle [95] mean that the formalism is

more cleanly integrated with other fields of physics. This is taken even further by the synthesis of quantum theory with statistical mechanics, making the path integral a natural candidate for describing stochastic quantum processes (in fact, the path integral can heuristically be interpreted as the analytic continuation of the Wiener integral for Brownian motion) [69]. The utility of the path integral is further apparent in its widespread application to describing non-perturbative corrections [74], novel Hamiltonians [70] and computer simulations [80].

In the context of this thesis, the path integral has a critical (although somewhat perverse) role in deriving the central results. The remainder of this section will be devoted to deriving the path integral, along with a few explicit calculations essential to later derivations.

2.1.1 Trotter Splitting

Consider a particle's path between two points x_i and x_f . This will be described (in co-ordinate space) by the propagator $U(x_f, t_f; x_i, 0) = \langle x_f, t_f | \hat{U}(t_f) | x_i, 0 \rangle$. To begin with, we will consider only time independent Hamiltonians, where $\hat{U}(t) = e^{-\frac{it}{\hbar} \hat{H}}$. Using the semi-group property of the propagator, the evolution can be partitioned into two sections

$$\langle x_f, t_f | \hat{U}(t_f) | x_i, 0 \rangle = \langle x_f, t_f | \hat{U}(t_f - \tau) \hat{U}(\tau) | x_i, 0 \rangle. \quad (2.1)$$

In the co-ordinate representation, the amplitude to reach x_f from x_0 must now sum over all possible values of the point x_τ

$$\langle x_f, t_f | \hat{U}(t_f - \tau) \hat{U}(\tau) | x_i, 0 \rangle = \int dx_\tau \langle x_f, t_f | \hat{U}(t_f - \tau) | x_\tau, \tau \rangle \langle x_\tau, \tau | \hat{U}(\tau) | x_i, 0 \rangle$$

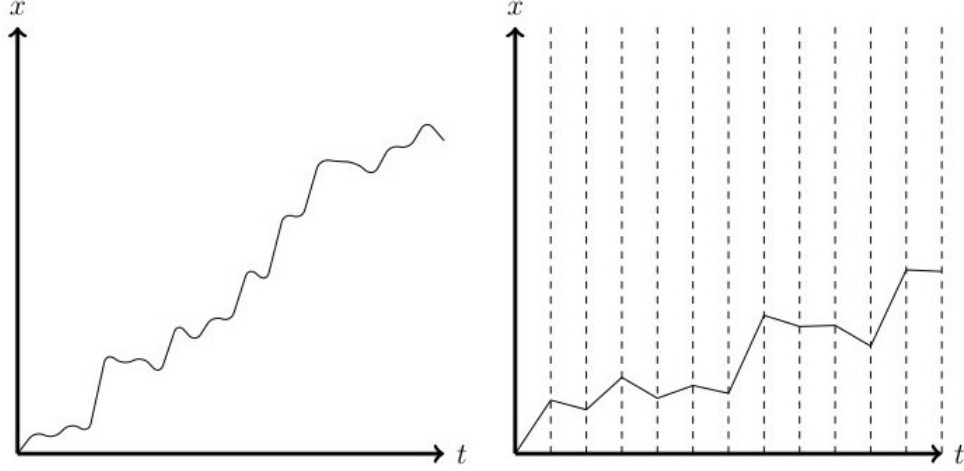


Figure 2.1: One dimensional evolution, where propagation is continuous in the left panel, and sliced into discrete sections on the right panel.

$$= \int dx_\tau \ U(x_f, t_f; x_\tau, \tau) U(x_\tau, \tau; x_i, 0) \quad (2.2)$$

This splitting process can be continued indefinitely, allowing one to decompose the propagator with N equally-spaced time slices of width Δ such that $t_f = N\Delta$. A graphical example of this is shown in Fig. 2.1. The propagator after slicing is then represented by

$$\hat{U}(t) = \prod_{j=0}^N e^{-\frac{i\Delta}{\hbar} \hat{H}}. \quad (2.3)$$

Evaluating this propagator in the coordinate basis is problematic, as \hat{H} is a sum of noncommuting operators for the kinetic energy \hat{T} and potential \hat{V} . Ideally, the infinitesimal propagator would be split into a product $\hat{U}(\Delta) \approx e^{-\frac{i\Delta}{\hbar} \hat{T}} e^{-\frac{i\Delta}{\hbar} \hat{V}}$ to allow for a direct calculation in the coordinate basis. The degree of approximation involved in splitting the Hamiltonian operator in this way depends on the slice width Δ . In the limit of zero width we have the identity

$$\lim_{N \rightarrow \infty} \left[\left(e^{-\frac{it_f}{N\hbar}(\hat{T} + \hat{V})} \right)^N - \left(e^{-\frac{it_f}{N\hbar}\hat{T}} e^{-\frac{it_f}{N\hbar}\hat{V}} \right)^N \right] = 0. \quad (2.4)$$

Proving this statement is not hard, but requires a slight detour. As a first step, note that the geometric series

$$\sum_{n=0}^N \left(\frac{Y}{X} \right)^n = \frac{1 - \left(\frac{Y}{X} \right)^{N+1}}{1 - \left(\frac{Y}{X} \right)} \quad (2.5)$$

can be rearranged to give

$$X^{N+1} - Y^{N+1} = (X - Y) \sum_{n=0}^N X^{(N-n)} Y^{n+1}. \quad (2.6)$$

We can use this identity to prove Eq.(2.4) with the substitution $X = e^{-\frac{i\Delta}{\hbar}(\hat{T} + \hat{V})}$ and $Y = e^{-\frac{i\Delta}{\hbar}\hat{T}} e^{-\frac{i\Delta}{\hbar}\hat{V}}$. Additionally, we will consider these operators as matrices and invoke the following generic inequalities for matrix norms¹ [96]:

$$|A + B| \leq |A| + |B| \quad (2.7)$$

$$|AB| \leq |A| |B| \quad (2.8)$$

$$|\exp(A)| \leq \exp(|A|). \quad (2.9)$$

These properties imply that X and Y simultaneously obey the inequality

$$|X|, |Y| \leq \exp \left(\frac{-it_f}{N\hbar} \left(|\hat{T}| + |\hat{V}| \right) \right) \equiv a^{\frac{1}{N}}. \quad (2.10)$$

¹Matrix norms are not uniquely defined, but they map a matrix to a real number and have a few generic properties. In this context, the norm $|A|$ can be thought of as a shorthand for the expectation of the operator between two arbitrary states $\langle \alpha | \hat{A} | \beta \rangle$.

Applying this to Eq. (2.6) yields a new inequality:

$$|X^N - Y^N| \leq N a^{\frac{N-1}{N}} |X - Y| \quad (2.11)$$

The behaviour of $X - Y$ in the large N limit can be ascertained using the Taylor expansion of $f(\alpha) = e^{\alpha \hat{T}} e^{-\alpha(\hat{T} + \hat{V})} e^{\alpha \hat{V}}$ about 0:

$$f(0) = 1 \quad (2.12)$$

$$f'(0) = 0 \quad (2.13)$$

$$f''(0) = [\hat{V}, \hat{T}] \quad (2.14)$$

$$f(\alpha) = 1 + \frac{\alpha^2}{2} [\hat{V}, \hat{T}] + \mathcal{O}(\alpha^3) \quad (2.15)$$

which implies

$$X - Y = \frac{t_f^2}{2N^2 \hbar^2} [\hat{V}, \hat{T}] + \mathcal{O}\left(\frac{1}{N^3}\right). \quad (2.16)$$

Inserting this into Eq. (2.11) demonstrates that to leading order in N

$$\left| \left(e^{-\frac{i\Delta}{\hbar}(\hat{T} + \hat{V})} \right)^N - \left(e^{-\frac{i\Delta}{\hbar}\hat{T}} e^{-\frac{i\Delta}{\hbar}\hat{V}} \right)^N \right| \leq \frac{1}{N} \left(\frac{t_f^2}{2\hbar^2} \right) a^{\frac{N-1}{N}}. \quad (2.17)$$

In the large N limit the bound becomes an equality and we obtain Eq.(2.4). We may therefore split the individual terms in Eq.(2.3) to identify the propagator as

$$\hat{U}(t) = \lim_{N \rightarrow \infty} \prod_{j=1}^N e^{-\frac{i\Delta}{\hbar}\hat{T}} e^{-\frac{i\Delta}{\hbar}\hat{V}}. \quad (2.18)$$

The process of dividing the propagator in order to express it as a product of eigen-operators of the position and momentum bases is known as Trotter splitting [97].

It is a critical step in deriving the path integral, and remains valid even when the Hamiltonian is time-dependent [50].

2.1.2 Expressing the propagator as a path integral

Having split the propagator, we can start to evaluate it in the coordinate space. We begin by inserting resolutions of the identity using the position basis at each of the N time slices

$$\begin{aligned}
U(x_f, t_f; x_i, 0) &= \lim_{N \rightarrow \infty} \langle x_f | \left(e^{-\frac{i\Delta}{\hbar} \hat{T}} e^{-\frac{i\Delta}{\hbar} \hat{V}} \right)^N | x_i \rangle \\
&= \lim_{N \rightarrow \infty} \int dx_1 \dots dx_{N-1} \left[\langle x_f | e^{-\frac{i\Delta}{\hbar} \hat{T}} e^{-\frac{i\Delta}{\hbar} \hat{V}} | x_{N-1} \rangle \dots \langle x_1 | e^{-\frac{i\Delta}{\hbar} \hat{T}} e^{-\frac{i\Delta}{\hbar} \hat{V}} | x_i \rangle \right] \\
&= \lim_{N \rightarrow \infty} \int dx_1 \dots dx_{N-1} \prod_{j=0}^{N-1} \langle x_{j+1} | e^{-\frac{i\Delta}{\hbar} \hat{T}} e^{-\frac{i\Delta}{\hbar} \hat{V}} | x_j \rangle. \tag{2.19}
\end{aligned}$$

In the last equality, $x_0 = x_i$ and $x_N = x_f$. These terminal states do not vary, and are not integrated over in the calculation of the propagator.

As the coordinate states are eigenbases of \hat{V} , we may partially evaluate each integrand:

$$\langle x_{j+1} | e^{-\frac{i\Delta}{\hbar} \hat{T}} e^{-\frac{i\Delta}{\hbar} \hat{V}} | x_j \rangle = \langle x_{j+1} | e^{-\frac{i\Delta}{\hbar} \hat{T}} | x_j \rangle e^{-\frac{i\Delta}{\hbar} V(x_j)}. \tag{2.20}$$

To proceed it is necessary to insert another identity resolved in the momentum basis, along with the overlap between position and momentum states $\langle p | x \rangle = \frac{1}{\sqrt{2\pi\hbar}} e^{ipx/\hbar}$,

$$\begin{aligned}
\langle x_{j+1} | e^{-\frac{i\Delta}{\hbar} \hat{T}} | x_j \rangle &= \int dp dp' \langle x_{j+1} | p' \rangle \langle p' | e^{-\frac{i\Delta}{\hbar} \hat{T}} | p \rangle \langle p | x_j \rangle \\
&= \int dp dp' \langle x_{j+1} | p' \rangle \langle p | x_j \rangle \delta(p - p') e^{-\frac{i\Delta p^2}{2m\hbar}}
\end{aligned}$$

$$\begin{aligned}
&= \frac{1}{2\pi\hbar} \int dp \, e^{-\frac{i\Delta p^2}{2m\hbar}} e^{ip(x_{j+1}-x_j)/\hbar} \\
&= \sqrt{\frac{m}{2\pi i\hbar\Delta}} e^{\frac{im\Delta}{2\hbar}(x_{j+1}-x_j)^2}.
\end{aligned} \tag{2.21}$$

Inserting Eqs. (2.20) & (2.21) into Eq. (2.19), we obtain (with a slight rearrangement) the path integral expression for the propagator:

$$\begin{aligned}
U(x_f, t_f; x_i, 0) &= \lim_{N \rightarrow \infty} \int dx_1 \dots dx_{N-1} \left(\frac{m}{2\pi i\hbar\Delta} \right)^{N/2} \\
&\times \exp \left[\frac{i\Delta}{\hbar} \sum_{j=0}^{N-1} \left[\frac{m}{2} \left(\frac{x_{j+1} - x_j}{\Delta} \right)^2 - V(x_j) \right] \right]
\end{aligned} \tag{2.22}$$

Very often, this expression is compactified with the following notation:

$$\lim_{N \rightarrow \infty} \Delta \sum_{j=0}^{N-1} \left[\frac{m}{2} \left(\frac{x_{j+1} - x_j}{\Delta} \right)^2 - V(x_j) \right] \rightarrow \int_0^{t_f} d\tau \left[\frac{m}{2} \left(\frac{dx}{d\tau} \right)^2 - V(x) \right] = S \tag{2.23}$$

$$\lim_{N \rightarrow \infty} \int dx_1 \dots dx_{N-1} \left(\frac{m}{2\pi i\hbar\Delta} \right)^{N/2} \rightarrow \int \mathcal{D}x \tag{2.24}$$

put together, we obtain the Feynman-Kac propagator [49]:

$$U(x_f, t_f; x_i, 0) = \int_{x_i}^{x_f} \mathcal{D}x(t) e^{\frac{iS}{\hbar}} \tag{2.25}$$

The integrand on the right hand side of Eq. (2.23) is immediately identifiable as the Lagrangian [19], confirming Dirac's original assertion that the path integral *is* the Lagrangian formulation of quantum mechanics.

2.1.3 Time-Dependent Propagator

While the derivation above was carried out with a time independent Hamiltonian, Eq. (2.25) also holds for a time dependent Hamiltonian [50]. This can be established by showing the wavefunction

$$\psi(x, t) = \int dx' U(x, t; x', t') \psi(x', t') \quad (2.26)$$

satisfies the time-dependent Schrödinger equation using Eq.(2.25) [96]. To prove this, we take the infinitesimal propagator between $t - \Delta$ and t in the form of Eq. (2.22):

$$U(x, t; x', t - \Delta) = \left(\frac{m}{2\pi i \hbar \Delta} \right)^{1/2} \exp \left[\frac{im}{2\hbar \Delta} (x - x')^2 - \frac{i\Delta}{\hbar} V(x, t - \Delta) \right] \quad (2.27)$$

where now $V(x, t)$ is now allowed to be time-dependent. Inserting this into Eq.(2.26) with the substitution $x' = y + x$ we obtain

$$\psi(x, t) = \left(\frac{m}{2\pi i \hbar \Delta} \right)^{1/2} \int dy \exp \left[\frac{im}{2\hbar \Delta} y^2 - \frac{i\Delta}{\hbar} V(y + x, t - \Delta) \right] \psi(y + x, t - \Delta). \quad (2.28)$$

Now we Taylor expand about x, t . Terms of order Δ^2 and higher will be neglected, while terms linear in y will integrate to zero and may also be dropped. The result is:

$$\begin{aligned} \psi(x, t) &= \left(\frac{m}{2\pi i \hbar \Delta} \right)^{1/2} \left(1 - \frac{i\Delta}{\hbar} V(x, t) \right) \\ &\times \int dy \exp \left[\frac{im}{2\hbar \Delta} y^2 \right] \left(\psi(x, t) - \Delta \dot{\psi}(x, t) + \frac{1}{2} y^2 \frac{d^2}{dx^2} \psi(x, t) \right) \end{aligned} \quad (2.29)$$

which is simply a Gaussian integral. Performing this yields

$$\psi(x, t) = \psi(x, t) - \frac{i\Delta}{\hbar} V(x, t) \psi(x, t) - \Delta \dot{\psi}(x, t) + \frac{i\hbar \Delta}{2m} \frac{d^2}{dx^2} \psi(x, t) + \mathcal{O}(\Delta^2). \quad (2.30)$$

After slight rearrangement we obtain the time-dependent Schrödinger equation,

$$i\hbar\dot{\psi}(x,t) = -\frac{\hbar^2}{2m}\frac{d^2}{dx^2}\psi(x,t) + V(x,t)\psi(x,t) \quad (2.31)$$

demonstrating that the path integral in Eq.(2.22) accurately describes the propagator even for a time-dependent Hamiltonian.

Aside: The Measure of a Quantum Path Integral

The adoption of functional integral notation in Eq. (2.25) is convenient, but duplicitous. Path integrals can be described as the continuous limit of a discrete lattice approximation, but this is not a guarantee that the limit exists. Indeed, the principal sin of this notation is concealment of the fact that $\mathcal{D}x$ is *not* a countably additive measure. Specifically, the measure is only finitely additive² over the paths [98], meaning that even if one were to take the lattice regularised version of the propagator in Eq.(2.22) and calculate each conditionally convergent integral before taking the limit, there is no guarantee that the result is convergent, let alone correct. Convergent integrals in a finitely additive measure occur due to precisely tuned cancellations in the divergent contributions to the integral, and this renders the result intrinsically dependent on the manner in which the limit is taken.

This problem extends beyond mere rigour- there are a great many Hamiltonians (e.g. electromagnetic potentials) where the correct Schrödinger equation is only derivable from the path integral with a specific discretisation [50]. This is somewhat

²The difference between a countable and finite additive measure can be summarised as follows- For a finite number of paths, the measure on each individual path adds up to the measure over all paths. Countable additivity is the stronger condition that this additive property for subsets of paths is retained in the limit of an infinite number of trajectories. For the Feynman path integral, one could choose a finite number of paths and define a measure that satisfies additivity, but it is impossible to fulfill this in the limit of infinite paths due to the factor of i attached to each path. This causes the measure to fluctuate and subsets of a collection of paths can have a larger measure than the set they belong to, breaking the entire notion of a measure.

analogous to the branching between the Ito and Stratonovich stochastic calculi, but in the case of the path integral there is only one correct choice of discretisation to maintain consistency with quantum mechanics.

Naturally, there have been a great many attempts to resolve this issue, and set the path integral on firmer mathematical foundations. The most productive early efforts stemmed from the observation that the Schrödinger equation is an analytic continuation of the diffusion equation [99]. This led to the proposal by Gel'fand³ and Yaglom that Feynman's path integral could be constructed as the analytic continuation of a (rigorous) Wiener process, with complex diffusion coefficient σ . Taking the limit $\text{Re}\sigma \rightarrow 0$ would then recover the quantum mechanical path integral [101]. Unfortunately, this development was scotched by later work which demonstrated that the Wiener measure is only countably additive when $\text{Re}\sigma > 0$, and is otherwise only finitely additive [102].

The path integral is not the only formulation of quantum dynamics with measure problems. The phase space formalism, where wavefunctions are replaced by the Wigner function [103,104] suffers from a lack of a true measure. In that case however, it is non-negativity rather than countable additivity that is missing. Both representations share the feature that they obscure the underlying non-commutative algebra, and attempt to “classicalise” the quantum formalism in different ways. The path integral takes a weighted sum of classical trajectories that potentially diverge, and the phase space representation is classical statistical dynamics with negative probabilities. To my mind, this seems to emphasise the impossibility of constructing a well-behaved quantum theory out of classical behaviours, despite the manifold historical attempts to do so.

Amusingly, given the issues bedeviling the path integral all lie in its continuous

³A mathematician with form when it comes to rigourising the innovations of theoretical physicists. The justification behind the bra-ket formalism requires what is known as either a rigged Hilbert space or Gel'fand triple [100].

limit, computer simulations relying on this method have exactly the same level of reliability as any other more rigorous method. This is due to the fact that numerics are implemented in a fundamentally discrete manner. In one sense this is reassuring, however it touches on the separate issue that *any* finite dimensional representation of quantum mechanics violates the Ehrenfest theorems [105]. This is particularly troubling, as a great deal of quantum mechanical proofs rely on precisely the opposite condition. Two previously encountered examples include the Trotter splitting formula and the Lindblad equation, both of which are derived on the presumption of bounded operators.

Finally, it is worth remarking on where genuine progress has been made on this problem. While the configuration path integral in Eq.(2.25) remains troublesome, the path integral formulated in phase space has been made consistent for the class of polynomial Hamiltonians [106]. This approach has the added bonus that its regularisation also involves a metric. While this technicality may initially appear inconsequential, it provides a geometric quantisation procedure⁴ that allows quantum mechanics to be expressed in a truly covariant, coordinate free form [107]. While there is still a question over the veracity of the path integral in general, for the class of Hamiltonians we will employ it is perfectly legitimate to use them.

2.1.4 Semi-Classical Approximation

While the path integral presents an appealing interpretation for quantum mechanics, actually performing calculations with it is far from straightforward. Functional integrals of the type seen in Eq. (2.25) are exactly solveable only for Lagrangians which are at most quadratic in their variables. To circumvent this problem, we make a change of variables $x(t) = \bar{x}(t) + y(t)$, where $\bar{x}(t)$ is the classical path that solves the

⁴The subtle argument for why one is ordinarily forced to quantise in Cartesian coordinates is that the same Hamiltonian function can represent a multiplicity of physical systems via a canonical transformation.

Euler-Lagrange equation and $y(t)$ are the fluctuations around it. This immediately imposes the constraint that $y(t)$ must be zero at the terminals of the path. Each of the integration variables x_j are just a translation of y_j , and each change of variable has a Jacobian of 1. Taylor expanding the action around the classical path yields

$$S = S_{\text{cl}} + \int_0^{t_f} dt \frac{\delta S}{\delta x(t)} \Big|_{\bar{x}} y(t) + \frac{1}{2} \iint_0^{t_f} dt dt' y(t) \frac{\delta^2 S}{\delta x(t) \delta x(t')} \Big|_{\bar{x}} y(t') + \dots \quad (2.32)$$

and for a Lagrangian of the form $L = \frac{1}{2}m\dot{x}^2 - V(x)$, the first two functional derivatives are

$$\frac{\delta S}{\delta x(t)} = V'(x(t)) - m\ddot{x}(t) \quad (2.33)$$

$$\frac{\delta^2 S}{\delta x(t) \delta x(t')} = -\delta(t - t') \left[\frac{\partial^2}{\partial t^2} + V''(x(t)) \right]. \quad (2.34)$$

Importantly, Eq. (2.33) evaluated on the classical path is, by definition, zero. The semi-classical approximation assumes that the quantum fluctuations are small, such that the higher order terms now shown in Eq.(2.32) may be neglected. The propagator can then be expressed as:

$$U(x_f, t_f; x_i, 0) = e^{i\frac{S_{\text{cl}}}{\hbar}} \tilde{U}(0, t_f; 0, 0). \quad (2.35)$$

With $\tilde{U}(0, t_f; 0, 0)$ providing a fluctuating factor:

$$\tilde{U}(0, t_f; 0, 0) = \int_{C\{0, t_f, 0, 0\}} \mathcal{D}x(t) \exp \left[\frac{i}{2\hbar} \int_0^{t_f} dt m \dot{x}^2(t) - V''(\bar{x}(t)) x^2(t) \right] \quad (2.36)$$

where $C\{0, t_f, 0, 0\}$ indicates an integral over all closed trajectories starting and ending at $x = 0$. This formulation allows the path integral to be used usefully as an approximation for any system, and as an **exact** result for quadratic Lagrangians⁵.

2.1.5 Propagator For The Free Particle

The simplest path integral propagator that can be evaluated is that of the free particle propagating from x_i to x_f in a time t_f . The classical action in this case is trivially $S_{\text{cl}} = \frac{(x_f - x_i)^2}{t_f^2}$, while the fluctuating factor is (in the discrete form):

$$\tilde{U}(0, t_f; 0, 0) = \lim_{N \rightarrow \infty} \int dx_1 \dots dx_{N-1} \left(\frac{m}{2\pi i \hbar \Delta} \right)^{N/2} \exp \left[\frac{im\Delta}{2\hbar} \sum_{j=0}^{N-1} \left(\frac{x_{j+1} - x_j}{\Delta} \right)^2 \right]. \quad (2.37)$$

The fluctuating factor will only depend on the endpoints of the trajectory, which can be demonstrated by considering a single integral in the product (say x_1), together with a factor $\left(\frac{m}{2\pi i \hbar \Delta} \right)^{1/2}$. This integral is Gaussian:

$$\int dx_1 \left(\frac{m}{2\pi i \hbar \Delta} \right)^{1/2} \exp \left[\frac{im}{2\Delta \hbar} [(x_2 - x_1)^2 + (x_1 - x_0)^2] \right] \quad (2.38)$$

and using the useful identity

$$\int_{-\infty}^{\infty} dy \sqrt{\frac{a}{\pi}} e^{-a(x_a - y)^2} \sqrt{\frac{b}{\pi}} e^{-b(y - x_b)^2} = \sqrt{\frac{ab}{\sqrt{a+b}}} \exp \left[-\frac{ab}{a+b} (x_a - x_b)^2 \right] \quad (2.39)$$

with $a = b = \left(\frac{m}{2i\hbar\Delta} \right)$, Eq. (2.38) can be evaluated

⁵Slightly less pragmatically, this form of the path integral makes the correspondence principle manifest. In the limit $\hbar \rightarrow 0$ we know via the method of stationary phase [50] that the only path that contributes to the propagator is exactly that of the classical action.

$$\left(\frac{m}{2\pi i\hbar(2\Delta)}\right)^{1/2} \exp\left[\frac{im}{2(2\Delta)\hbar}(x_2 - x_0)^2\right]. \quad (2.40)$$

Importantly, the result of this integration is of the same functional form as the initial integrand (with a rescaled Δ). Repeated application of Eq. (2.39) to Eq. (2.37) results in the j th integration being

$$\begin{aligned} \int_{-\infty}^{\infty} dx_j \sqrt{\frac{m}{2\pi i\hbar\Delta}} e^{-\frac{im}{2(\Delta)\hbar}(x_{j+1}-x_j)^2} \sqrt{\frac{im}{2(j\Delta)\hbar}} e^{-\frac{im}{2(j\Delta)\hbar}(x_j-x_i)^2} \\ = \sqrt{\frac{im}{2((j+1)\Delta)\hbar}} \exp\left[-\frac{im}{2((j+1)\Delta)\hbar}(x_{j+1}-x_0)^2\right]. \end{aligned} \quad (2.41)$$

Finally, after N integrations we obtain the fluctuating factor

$$\tilde{U}(0, t_f; 0, 0) = \lim_{N \rightarrow \infty} \left(\frac{m}{2\pi i\hbar N\Delta}\right)^{1/2} \left[\frac{im}{2\hbar t_f}(x_N - x_0)^2\right]_{x_0=x_N} = \sqrt{\frac{m}{2\pi i\hbar t_f}}. \quad (2.42)$$

The free-particle propagator is therefore

$$U(x_f, t_f; x_i, 0) = \sqrt{\frac{m}{2\pi i\hbar t_f}} \exp\left[\frac{im}{2\hbar t_f}(x_f - x_i)^2\right]. \quad (2.43)$$

2.1.6 The Harmonic Oscillator

Having demonstrated the calculability of the free-particle propagator, it is natural to turn to the canonical problem in any area of physics, the harmonic oscillator. This will have the Lagrangian:

$$L = \frac{m}{2}\dot{x}^2 - \frac{1}{2}g(t)x^2(t) - f(t)x(t) \quad (2.44)$$

In anticipation of its later use, we have attempted to use the most general for-

mulation possible (allowing time dependent frequencies and an arbitrary damping function). Consider first the fluctuating factor. Using Eq. (2.34) this is given by:

$$\tilde{U}(0, t_f; 0, 0) = \int_{C\{0, t_f, 0, 0\}} \mathcal{D}x(t) \exp \left[\frac{i}{2\hbar} \int_0^{t_f} dt m \dot{x}^2(t) - g(t) \right]. \quad (2.45)$$

Performing this integral requires a reversion to the lattice discretisation:

$$\tilde{U}(0, t_f; 0, 0) = \lim_{N \rightarrow \infty} \int dx_1 \dots dx_{N-1} \left(\frac{m}{2\pi i \hbar \Delta} \right)^{N/2} \exp \left[\frac{i}{2\hbar} \sum_{j=0}^{N-1} \left[\frac{m}{\Delta} (x_{j+1} - x_j)^2 - \Delta g_j x_j^2 \right] \right] \quad (2.46)$$

While it is possible to evaluate this integral in a variety of ways (exploiting the boundary conditions to express the coordinates as Fourier series for instance) [50], the most direct method is to represent the integral in matrix form:

$$\tilde{U}(0, t_f; 0, 0) = \lim_{N \rightarrow \infty} \left(\frac{m}{2\pi i \hbar \Delta} \right)^{N/2} \int d\mathbf{x} \exp \left[-\mathbf{x}^T \Sigma \mathbf{x} \right]. \quad (2.47)$$

Here \mathbf{x} is just the vector of coordinates x_j , while Σ is the matrix defined by:

$$\Sigma = -\frac{im}{2\Delta\hbar} \begin{bmatrix} 2 & -1 & 0 & \dots & 0 \\ -1 & 2 & -1 & \ddots & \vdots \\ 0 & -1 & 2 & \ddots & 0 \\ \vdots & \ddots & \ddots & \ddots & -1 \\ 0 & \dots & 0 & -1 & 2 \end{bmatrix} + \frac{i\Delta}{2\hbar} \begin{bmatrix} g_1 & 0 & \dots & \dots & 0 \\ 0 & \ddots & 0 & \ddots & \vdots \\ \vdots & 0 & \ddots & 0 & \vdots \\ \vdots & \ddots & 0 & \ddots & 0 \\ 0 & \dots & \dots & 0 & g_{N-1} \end{bmatrix}. \quad (2.48)$$

Σ is composed of a tridiagonal and diagonal matrix, each of size $(N-1) \times (N-1)$ due to the the boundary condition $x_0 = x_N = 0$. Once again, the integral is Gaussian, and will generate prefactor of $\frac{\pi^{(N-1)/2}}{\sqrt{|\Sigma|}}$ [50]. Rearranging, the expression for \tilde{U} is

$$\tilde{U}(0, t_f; 0, 0) = \lim_{N \rightarrow \infty} \left[\frac{m}{2\pi i \hbar} \frac{1}{\Delta C} \right]^{1/2} \quad (2.49)$$

$$C(t_f) = \left(\frac{2i\hbar\Delta}{m} \right)^{N-1} |\Sigma|. \quad (2.50)$$

At first glance calculating the fluctuating factor now appears hopeless, as C is a function of the determinant of an infinite (in the limit) matrix. In this special case however the structure of Σ makes evaluation possible even in the infinite size limit.

Consider the relationship between the determinants of submatrices of C . The determinant of the $j \times j$ submatrix determinant $|C|_j$ is equivalent to $|C(t = j\Delta)|$ and can be expressed recursively

$$|C|_{j+1} = \left(2 - \frac{\Delta^2}{m} g_{j+1} \right) |C|_j - |C|_{j-1}. \quad (2.51)$$

Defining $|F|_0 = 1$ and $|F|_{-1} = 0$, this recursion relation can be rearranged to give

$$\frac{|C|_{j+1} - 2|C|_j + |C|_{j-1}}{\Delta^2} = -\frac{g_{j+1}}{m} |C|_j. \quad (2.52)$$

In the limit of $\Delta \rightarrow 0$ (or equivalently $N \rightarrow \infty$), this starts to resemble a differential equation. We can make this a function of time using $\phi(t) = \Delta |C|_j$, and obtain:

$$\frac{d^2 \phi(t)}{dt^2} = -\frac{g(t)}{m} \phi(t). \quad (2.53)$$

The initial conditions for this differential equation follow on from the recursion relation definitions, i.e. $\phi(0) = 0$, $\phi'(0) = 1$. Consequently the fluctuation factor can be determined by solving the differential equation for $f(\tau)$ and evaluating it at t_f . The fluctuating factor is therefore

$$\tilde{U}(0, t_f; 0, 0) = \sqrt{\frac{m}{2\pi i \hbar \phi(t_f)}}. \quad (2.54)$$

For a simple harmonic oscillator, $g(t) = m\omega^2$, making Eq. (2.53) trivial to solve and yielding $\phi(t_f) = \frac{\sin(\omega t_f)}{\omega}$.

Having fixed the fluctuating factor, we may now proceed to calculating the classical action. In the case of $g(t) = m\omega^2$, solving the Euler-Lagrange equation gives the following equation of motion⁶

$$m\ddot{x}(t) = -m\omega^2 x(t) + f(t) \quad (2.55)$$

whose solution is required for the classical action for the system.

Solving this equation is not entirely trivial, but can be accomplished in a variety of ways (a Green's function approach is often used here). In the interest of novelty we shall take a slightly different approach, which to the best of my knowledge has not previously been applied to this problem. First we re-express Eq. (2.55) as a first order matrix equation

$$m\dot{X}(t) = mAX + F(t) \quad (2.56)$$

$$X = \begin{pmatrix} x(t) \\ \dot{x}(t) \end{pmatrix} \quad A = \begin{pmatrix} 0 & 1 \\ -\omega^2 & 0 \end{pmatrix} \quad F = \begin{pmatrix} 0 \\ f(t) \end{pmatrix}. \quad (2.57)$$

Solving this equation with the integrating factor $\exp(At)$ yields

$$X(t) = e^{-At} X(0) + \frac{1}{m} \int_0^t ds e^{-A(t-s)} f(s). \quad (2.58)$$

This solution can be recast by expanding the matrix exponentials. This first requires

⁶Its also possible to solve the equation of motion exactly when $g(t)$ is an integrable function [108, 109], but for our purposes it is sufficient to stick to a fixed frequency.

the evaluation of A^n :

$$A^n = \begin{cases} (-1)^{n/2} \omega^n \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} & n \text{ even} \\ (-1)^{(n-1)/2} \omega^{n-1} \begin{pmatrix} 0 & 1 \\ -\omega^2 & 0 \end{pmatrix} & n \text{ odd} \end{cases} \quad (2.59)$$

which can be used to rearrange the exponential expansion into odd and even terms

$$\begin{aligned} e^{-At} &= \sum_{n=0}^{\infty} \left\{ \frac{(At)^{2n}}{(2n)!} + \frac{(At)^{2n+1}}{(2n+1)!} \right\} \\ &= \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n)!} (\omega t)^{2n} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \frac{1}{\omega} \frac{(-1)^n}{(2n+1)!} (\omega t)^{2n+1} \begin{pmatrix} 0 & 1 \\ -\omega^2 & 0 \end{pmatrix} \\ &= \cos(\omega t) \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \sin(\omega t) \begin{pmatrix} 0 & \omega^{-1} \\ -\omega & 0 \end{pmatrix}. \end{aligned} \quad (2.60)$$

Substituting this into Eq. (2.58) we obtain:

$$\begin{aligned} \begin{pmatrix} x(t) \\ \dot{x}(t) \end{pmatrix} &= \cos(\omega t) \begin{pmatrix} x(0) \\ \dot{x}(0) \end{pmatrix} + \sin(\omega t) \begin{pmatrix} \dot{x}(0)/\omega \\ -\omega x(0) \end{pmatrix} \\ &+ \int_0^t ds \cos(\omega(t-s)) \begin{pmatrix} 0 \\ f(s) \end{pmatrix} + \frac{1}{m} \int_0^t ds \sin(\omega(t-s)) \begin{pmatrix} f(s)/\omega \\ 0 \end{pmatrix} \end{aligned} \quad (2.61)$$

A nice feature of this method is that once the equation of motion is obtained reading off the top row, there is a free consistency check that its derivative is equal to the bottom row. Using A and B for constants and grouping terms produces

$$x(t) = A \sin(\omega t) + B \sin(\omega(t_f - t)) + \frac{1}{m\omega} \int_0^t ds f(s) \sin(\omega(t - s)) \quad (2.62)$$

$$\dot{x}(t) = A\omega \cos(\omega t) - B\omega \cos(\omega(t_f - t)) + \frac{1}{m} \int_0^t ds f(s) \cos(\omega(t - s)). \quad (2.63)$$

Having obtained the equation of motion, we are now ready to calculate the classical action:

$$S_{\text{cl}} = \int_0^{t_f} dt \left(\frac{m}{2} \dot{x}(t)^2 - \frac{m}{2} \omega^2 x(t)^2 + f(t)x(t) \right). \quad (2.64)$$

Directly substituting Eq. (2.62) into the action would lead to a trigonometric nightmare from which there is no escape. Instead, integrate the kinetic term by parts [49]

$$\int_0^{t_f} dt \frac{m}{2} \dot{x}(t)^2 = \frac{m}{2} [x(t)\dot{x}(t)]_0^{t_f} + \frac{1}{2} \int_0^{t_f} dt (m\omega^2 x(t)^2 - f(t)x(t)) \quad (2.65)$$

which when inserted into the action reduces it to

$$S_{\text{cl}} = \frac{m}{2} [x(t)\dot{x}(t)]_0^{t_f} + \frac{1}{2} \int_0^{t_f} dt f(t)x(t). \quad (2.66)$$

The equation of motion for the oscillator from Eq. (2.62) may now be applied to Eq. (2.66). Before performing this substitution, some convenient definitions are

$$F_s = \frac{1}{m\omega} \int_0^{t_f} ds f(s) \sin(\omega(t_f - s)) \quad (2.67)$$

$$F_c = \frac{1}{m\omega} \int_0^{t_f} ds f(s) \cos(\omega(t_f - s)). \quad (2.68)$$

Setting $x(0) = x_i$ and $x(t_f) = x_f$ gives explicit values for A and B in Eq. (2.62)

$$A = \frac{x_f - F_s}{\sin(\omega t_f)} \quad (2.69)$$

$$B = \frac{x_i}{\sin(\omega t_f)} \quad (2.70)$$

which may be substituted into the term $[x(t)\dot{x}(t)]_0^{t_f}$ in Eq. (2.66):

$$\begin{aligned} [x(t)\dot{x}(t)]_0^{t_f} &= x_f \omega \left[\left(\frac{x_f - F_s}{\sin(\omega t_f)} \right) \cos(\omega t_f) - \left(\frac{x_i}{\sin(\omega t_f)} \right) + F_c \right] \\ &\quad - x_i \omega \left[\left(\frac{x_f - F_s}{\sin(\omega t_f)} \right) - \left(\frac{x_i}{\sin(\omega t_f)} \right) \cos(\omega t_f) \right]. \end{aligned} \quad (2.71)$$

By collecting terms in x_f and x_i this can be formulated as

$$[x(t)\dot{x}(t)]_0^{t_f} = \frac{\omega}{\sin(\omega t_f)} \left[(x_f^2 + x_i^2) \cos(\omega t_f) - 2x_f x_i \right. \quad (2.72)$$

$$\left. + x_f (F_c \sin(\omega t_f) - F_s \cos(\omega t_f)) + x_i F_s \right]. \quad (2.73)$$

The third term in this expression can be simplified further, noting that $F_c \sin(\omega t_f) - F_s \cos(\omega t_f)$ can be reduced via the sine compound angle identity

$$F_c \sin(\omega t_f) - F_s \cos(\omega t_f) = \frac{1}{m\omega} \int_0^{t_f} ds f(s) \sin(\omega s). \quad (2.74)$$

Applying this to Eq. (2.72), we obtain a final expression for the first term of the classical action

$$\frac{m}{2} [x(t)\dot{x}(t)]_0^{t_f} = \frac{m\omega}{2 \sin(\omega t_f)} \left[x_f^2 + x_i^2 \cos(\omega t_f) - x_f x_i \right.$$

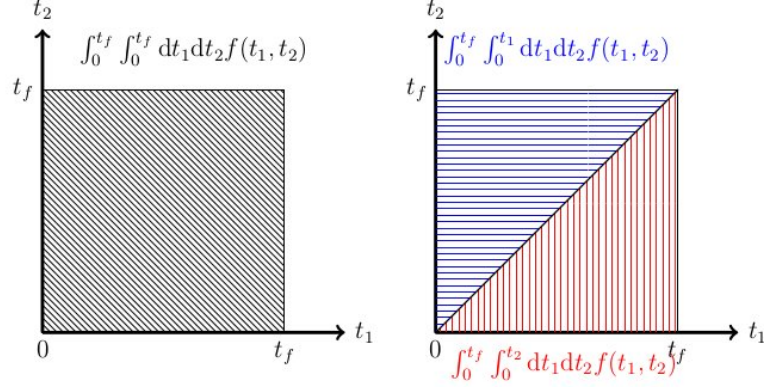


Figure 2.2: Geometric representation of limit swapping with hatching indicating the direction of the first integration

$$+\frac{x_f}{m\omega} \int_0^{t_f} ds f(s) \sin(\omega s) + \frac{x_i}{m\omega} \int_0^{t_f} ds f(s) \sin(\omega(t_f - s)) \Big]. \quad (2.75)$$

The second term in Eq.(2.66) may be expanded in the same manner as the first, yielding

$$\begin{aligned} \frac{1}{2} \int_0^{t_f} dt f(t)x(t) &= \frac{1}{2 \sin(\omega t_f)} \left[x_f \int_0^{t_f} ds f(s) \sin(\omega s) + x_i \int_0^{t_f} ds f(s) \sin(\omega(t_f - s)) \right. \\ &\quad - \frac{1}{m\omega} \int_0^{t_f} \int_0^{t_f} ds du f(s)f(u) \sin(\omega(t_f - s)) \sin(\omega u) \\ &\quad \left. + \frac{1}{m\omega} \int_0^{t_f} \int_0^s ds du f(s)f(u) \sin(\omega(s - u)) \sin(\omega t_f) \right]. \end{aligned} \quad (2.76)$$

At this point the terms involving multiple integrals require some reformulation. To achieve this, examine the term on the second line of Eq. (2.76). This is an integration over a square area, of the type $\int_0^{t_f} \int_0^{t_f} dt_1 dt_2 g(t_1, t_2)$ where g is some arbitrary function. Fig. 2.2 demonstrates how this is split up into two integrals. As a result, we have the equation

$$\begin{aligned}
\int_0^{t_f} \int_0^{t_f} dt_1 dt_2 g(t_1, t_2) &= \int_0^{t_f} \int_0^{t_1} dt_1 dt_2 g(t_1, t_2) + \int_0^{t_f} \int_0^{t_2} dt_1 dt_2 g(t_1, t_2) \\
&= \int_0^{t_f} \int_0^{t_2} dt_1 dt_2 (g(t_1, t_2) + g(t_2, t_1))
\end{aligned} \tag{2.77}$$

where the final equality is obtained by swapping the arguments t_1 and t_2 . Applying this to the square integration term gives

$$\begin{aligned}
&\frac{1}{m\omega} \int_0^{t_f} \int_0^{t_f} ds du f(s) f(u) \sin(\omega(t_f - s)) \sin(\omega u) = \\
&\int_0^{t_f} \int_0^s ds du f(s) f(u) \left[\sin(\omega(t_f - s)) \sin(\omega u) + \sin(\omega(t_f - u)) \sin(\omega s) \right].
\end{aligned} \tag{2.78}$$

If this term is now inserted back into eqn. (2.76), we obtain

$$\begin{aligned}
\frac{1}{2} \int_0^{t_f} dt f(t) x(t) &= \frac{1}{2 \sin(\omega t_f)} \left[x_f \int_0^{t_f} ds f(s) \sin(\omega s) + x_i \int_0^{t_f} ds f(s) \sin(\omega(t_f - s)) \right. \\
&\quad \left. - \frac{2}{m\omega} \int_0^{t_f} \int_0^s ds du f(s) f(u) \sin(\omega(t_f - s)) \sin(\omega u) \right]
\end{aligned} \tag{2.79}$$

where the last equality follows from the trigonometric compound angle identities.

Finally, adding together Eq. (2.75) and Eq. (2.79) gives the classical action:

$$\begin{aligned}
S_{\text{cl}} &= \frac{m\omega}{\sin(\omega t_f)} \left[\frac{1}{2} (x_f^2 + x_i^2) \cos(\omega t_f) - x_f x_i \right. \\
&\quad + \frac{x_f}{m\omega} \int_0^{t_f} ds f(s) \sin(\omega s) + \frac{x_i}{m\omega} \int_0^{t_f} ds f(s) \sin(\omega(t_f - s)) \\
&\quad \left. - \frac{1}{(m\omega)^2} \int_0^{t_f} \int_0^s ds du f(s) f(u) \sin(\omega(t_f - s)) \sin(\omega u) \right].
\end{aligned} \tag{2.80}$$

It took a little while, but it is now possible to state the general form for a propagator

under a quadratic Lagrangian⁷:

$$\begin{aligned}
U(x_f, t_f; x_i, 0) = & \sqrt{\frac{m\omega}{2\pi i\hbar \sin(\omega t)}} \exp \left\{ \frac{im\omega}{\hbar \sin(\omega t_f)} \left[\frac{1}{2} (x_f^2 + x_i^2) \cos(\omega t_f) - x_f x_i \right. \right. \\
& + \frac{x_f}{m\omega} \int_0^{t_f} ds f(s) \sin(\omega s) + \frac{x_i}{m\omega} \int_0^{t_f} ds f(s) \sin(\omega(t_f - s)) \\
& \left. \left. - \frac{1}{(m\omega)^2} \int_0^{t_f} \int_0^s ds du f(s)f(u) \sin(\omega(t_f - s)) \sin(\omega u) \right] \right\} \quad (2.81)
\end{aligned}$$

2.1.7 The Density Matrix as a Path Integral

The discussion thus far has centred on propagators for pure quantum states, restricting the formalism to systems lacking many of the interesting properties we may wish to study. The principal goal of this section is an incorporation of temperature and thermodynamics into the path integral representation. If we consider a system in thermal equilibrium, then a statistical uncertainty is introduced to the initial state of the system, and hence to its potential final states. Path integrals offer a compelling description of statistical phenomena and the density matrix may also be cast in this form.

First, note that the thermal density matrix may be expressed as a propagator in *imaginary time*:

$$\hat{U}(-i\hbar\beta) = e^{-\beta\hat{H}} = Z_\beta \hat{\rho}_\beta \quad (2.82)$$

⁷While Eq.(2.81) is correct, the prefactor is ambiguous. A naive implementation of this propagator will only give the correct result *half* the time, and a careful analysis reveals that there is a sign change in the propagator after each classical time period of the system [110] .

where $\hat{\rho}_\beta$ is the thermal density matrix and Z_β is its partition function. Here, β is a parameter that we can split into arbitrarily many pieces Δ , with $\Delta = \hbar\beta/N$. Now the density matrix is expressible as

$$\rho_\beta = \frac{1}{Z_\beta} \left(e^{-\frac{\Delta}{\hbar} H} \right)^N. \quad (2.83)$$

Performing the same Trotter splitting as before yields:

$$\rho_\beta(x, x') = \frac{1}{Z_\beta} \lim_{N \rightarrow \infty} \int dx_1 \dots dx_{N-1} \left(\frac{m}{2\pi i \hbar \Delta} \right)^{N/2} \exp \left[-\frac{\Delta}{\hbar} \sum_{j=0}^{N-1} \left[\frac{m}{2} \left(\frac{x_{j+1} - x_j}{\Delta} \right)^2 + V(x_j) \right] \right] \quad (2.84)$$

using $x = x_0$, $x' = x_N$. The shorthand for this integral is

$$\rho(x, x') = \frac{1}{Z_\beta} \int \mathcal{D}\bar{x}(\tau) e^{-\frac{1}{\hbar} S^E} \quad (2.85)$$

where S^E is the *Euclidean action*

$$S^E = \int_0^{\hbar\beta} d\tau \left(\frac{1}{2} m \dot{\bar{x}}^2(\tau) + V(\bar{x}(\tau)) \right). \quad (2.86)$$

There is a simple way to relate the Euclidean action to Lagrangian action, allowing all the results derived for propagators to be used for the density matrix. First, performing a Wick rotation $t \rightarrow -i\tau$ on the propagator:

$$U(x_f, t_f; x_0, 0) \rightarrow U(x_f; x_0; -i\tau_f) = \int \mathcal{D}x(-i\tau_f) e^{\frac{i}{\hbar} \bar{S}} \quad (2.87)$$

$$\bar{S} = -i \int_0^{\tau_f} L(-i\tau) d\tau. \quad (2.88)$$

After substituting $\bar{x}(\tau) = x(-i\tau)$ and $\tau_f = \hbar\beta$ we obtain:

$$\bar{S} = i \int_0^{\hbar\beta} d\tau \frac{1}{2} m \dot{\bar{x}}^2(\tau) + V(\bar{x}(\tau)) = iS^E \quad (2.89)$$

$$U(x, -i\hbar\beta; x') = \int \mathcal{D}\bar{x}(\tau) e^{-\frac{1}{\hbar} S^E} = Z_\beta \rho_\beta(x; x') \quad (2.90)$$

This confirms that the correspondence between the propagator and density matrix operators under a Wick rotation also holds in the path integral formulation. Using this fact allows the results for any calculated propagator to be used for the canonical density matrix with the same Hamiltonian - all time arguments are simply replaced by $-i\tau$, ($-i\hbar\beta$ for the final time). The canonical density matrix for a general quadratic Lagrangian is therefore:

$$\begin{aligned} \rho(x; x') = \frac{1}{Z_\beta} \sqrt{\frac{m\omega}{2\pi\hbar \sinh(\omega t)}} \exp \left\{ -\frac{m\omega}{\hbar \sinh(\omega\hbar\beta)} \left[\frac{1}{2} (x^2 + x'^2) \cosh(\omega\hbar\beta) - xx' \right. \right. \\ \left. \left. - \frac{x}{m\omega} \int_0^{\hbar\beta} d\tau f(\tau) \sinh(\omega\tau) - \frac{x'}{m\omega} \int_0^{\hbar\beta} d\tau f(\tau) \sinh(\omega(\hbar\beta - \tau)) \right] \right\} \quad (2.91) \end{aligned}$$

2.1.8 The Influence Functional

The path integrals considered so far have dealt purely with one-particle systems, but one of the main utilities of path integrals are their ability to re-express a many particle problem in terms of a modified one-body equation. This is done via another Feynman innovation, the *influence functional* [57].

Consider two subsystems Q and X respectively characterised by collective coordinates q and x , with an interaction $H_I(q, x)$. The total Hamiltonian is described

by

$$H_{\text{tot}}(q, x) = H_Q(q) + H_X(x) + H_I(q, x). \quad (2.92)$$

Let us imagine that we are only interested in the dynamics of one of the subsystems (say, q). The expectation of an operator \hat{A} acting only on the Q subsystem is

$$\langle \hat{A} \rangle = \int \rho(q, x; q', x'; t) A(q, q') \delta(x - x') dq dq' dx dx'. \quad (2.93)$$

This expression can be simplified by defining a *reduced* density matrix $\hat{\rho}_Q(t)$ which describes subsystem Q

$$\rho_Q(q; q'; t) = \int \rho(q, x; q', x'; t) \delta(x - x') dx dx'. \quad (2.94)$$

So far, this is simply a restatement of the fundamental properties of the density matrix given in section 1.2. Additionally, when we incorporate time evolution, the density matrix at time t_f is

$$\rho(q, x; q', x'; t_f) = \int dq_0 dq'_0 dx_0 dx'_0 U(q, x; q_0, x'_0; t_f) \rho_0(q_0, q'_0; x_0, x'_0) U^\dagger(q', x'; q'_0, x'_0; t_f). \quad (2.95)$$

Notice that for density matrices there are two propagators acting on the unprimed and primed coordinates at either side of the density matrix, which can be interpreted as forward and reversed time trajectories respectively [91].

If we insert the path integral representation for the propagators we obtain:

$$\rho(q, x; q', x'; t_f) = \int \mathcal{D}Q \mathcal{D}X e^{i \frac{S_{\text{tot}}}{\hbar}} \rho(q_0, q'_0; x_0, x'_0; t_f)$$

where in the interests of concision we have made the abbreviations:

$$dq_0 dq'_0 \mathcal{D}q(t) \mathcal{D}q'(t) = \mathcal{D}Q \quad (2.96)$$

$$dx_0 dx'_0 \mathcal{D}x(t) \mathcal{D}x'(t) = \mathcal{D}X \quad (2.97)$$

$$\begin{aligned} S_{\text{tot}} = & S_Q [q(t)] - S_Q [q'(t)] + S_X [x(t)] \\ & - S_X [x'(t)] + S_I [q(t), x(t)] - S_I [q'(t), x'(t)]. \end{aligned} \quad (2.98)$$

$S_{Q/X}$ are the actions derived from the isolated Q and X subsystem Hamiltonians, while S_I is the component due to the coupling H_I . This last equality is somewhat misleading, given the action is a functional of both the coordinates and their time derivatives. The functional arguments should be therefore be thought of purely as labels denoting whether a particular component of the action is due to the forward or backward propagator trajectories.

Usually when calculating dynamical properties of the reduced system, it is assumed that the density matrix is initially in a *product state*, that is:

$$\rho_0 = \rho_Q(q_0; q'_0) \rho_X(x_0; x'_0) \quad (2.99)$$

where the time coordinate in the initial density matrices is implicitly 0. Partitioning the initial density matrix is equivalent to stipulating that the interaction between the systems is only turned on at time 0_+ . As discussed in the introduction, this is an artificial constraint which precludes strong couplings between the two systems, and later sections shall proceed without it.

For an initial product state, the reduced density matrix is

$$\rho_Q(q; q'; t) = \int \mathcal{D}q(t) \mathcal{D}q'(t) dq_0 dq'_0 \mathcal{F}[q(t), q'(t)] \rho_Q(q_0; q'_0) \exp(i/\hbar [S_Q[q(t)] - S_Q[q'(t)]]) \quad (2.100)$$

$$\begin{aligned} \mathcal{F}[q(t), q'(t)] = & \int dx_0 dx'_0 dx \mathcal{D}x(t) \mathcal{D}x'(t) \left[\rho_X(x_0; x'_0) \right. \\ & \left. \times \exp(i/\hbar [S_X[x(t)] - S_X[x'(t)] + S_I[q(t), x(t)] - S_I[q'(t), x'(t)]]) \right]. \end{aligned} \quad (2.101)$$

Here $\mathcal{F}(q, q')$ is the *influence functional*. This object explicitly integrates out the X system, leaving it a pure function of the Q system coordinates. If the influence functional is expressed as a complex phase

$$F[q(t), q'(t)] = \exp\left(\frac{i}{\hbar} \Phi[q(t), q'(t)]\right) \quad (2.102)$$

then it is possible to define an *effective* density-matrix propagator for the evolution of the Q system:

$$U_Q[q(t), q'(t)] = \exp(i/\hbar [S_Q[q(t)] - S_Q[q'(t)] + \Phi[q(t), q'(t)]]) \quad (2.103)$$

$$\rho_Q(q; q'; t) = \int \mathcal{D}q(t) \mathcal{D}q'(t) dq_0 dq'_0 U_Q[q(t), q'(t)] \rho_Q(q_0; q'_0) \quad (2.104)$$

Furthermore, if one is able to disentangle U_Q into a product of the form

$$U_Q[q(t), q'(t)] = \tilde{U}_Q[q(t)] \tilde{U}_Q^\dagger[q'(t)] \quad (2.105)$$

then an effective Hamiltonian can also be defined, capturing exactly the dynamics of the Q system, but without any reference to the X system it is interacting with. This is the power of the influence functional, as it allows for the mapping of an

interacting subsystem to an isolated system with a modified Hamiltonian. In the context of open systems, the dimensionality of the environment is incomparably large as compared to the system of interest. Being able to use the influence functional to characterise the environmental effect on the open system is highly desirable (even if only for numerical efficiency). Unfortunately, there is no such thing as a free lunch, and the influence functional is no exception. Finding an explicit expression for even the simplest environmental models requires extensive work. Nevertheless, it *is* possible, and chapter 3 details explicitly the calculation of a generalised influence functional.

2.2 Hubbard-Stratonovich Transformation

In this section we will explore some useful theorems in statistics, using them to eventually reformulate influence functionals in such a way that the path integral prescription can be reversed. All of this is in the service of the ultimate aim of deriving an effective equation of motion for the Q system without reference to X . In our case, this ultimately requires the application of a Hubbard-Stratonovich (HS) transformation in two time dimensions. This in turn necessitates a short diversion into the mathematics of Gaussian processes.

2.2.1 Real Gaussian processes

Consider a collection of Gaussianly distributed real random variables z_i described by the set $\{z_i\}$. We can associate to each z_i an ordering parameter t_i . This is then a multivariate Gaussian process whose joint distribution can be expressed as:

$$P(z_1, t_1; \dots; z_N, t_N) = C \exp \left[-\frac{1}{2} \sum_i^N a_{ij} (z_i - m_i)(z_j - m_j) \right]$$

$$= C \exp \left[-\frac{1}{2}(\mathbf{z} - \mathbf{m})^T A(\mathbf{z} - \mathbf{m}) \right] \quad (2.106)$$

where C is the normalisation and $m_i = \langle z_i \rangle$. By ordering the N random variables by the parameter t_i , this Gaussian process can be interpreted as the probability for the trajectory of a single random variable over time. Before this can be used productively, it is worth checking that the usual Gaussian distribution properties apply. For instance, the correlation function between two given variables is:

$$\begin{aligned} \langle (z_j - m_j)(z_k - m_k) \rangle &= \int d\mathbf{z} P(\mathbf{z}, t) (z_j - m_j)(z_k - m_k) \\ &= C \int d\mathbf{x} x_j x_k \exp \left[-\frac{1}{2} \mathbf{x}^T A \mathbf{x} \right] \end{aligned} \quad (2.107)$$

using $\mathbf{z}_i - \mathbf{m}_i = \mathbf{x}_i$ in the last equality. Eq. (2.107) is most easily resolved using the eigenvectors of the matrix A [111]: $A\mathbf{e}_\lambda = a_\lambda \mathbf{e}_\lambda$, $A = \sum_\lambda a_\lambda \mathbf{e}_\lambda \mathbf{e}_\lambda^T$. A is a symmetric matrix, so by the spectral theorem the eigenvectors have an orthogonal basis. A new variable $y_\lambda = \mathbf{x}^T \mathbf{e}_\lambda = \sum_i x_i e_{\lambda i}$, can be defined where the e_{ij} are elements in an orthonormal matrix constructed from the eigenvectors (each row/column is an eigenvector). The inverse relation is easily found from this orthogonality as $x_i = \sum_\lambda y_\lambda e_{\lambda i}$. The Jacobian for this transformation is:

$$J = \left| \frac{\partial x_i}{\partial y_\lambda} \right| = |e_{\lambda i}| = 1 \quad (2.108)$$

and the integrand for the correlation function is now given by:

$$x_j x_k \exp \left[-\frac{1}{2} \mathbf{x}^T A \mathbf{x} \right] = \sum_{\lambda_1 \lambda_2} y_{\lambda_1} y_{\lambda_2} e_{\lambda_1 j} e_{\lambda_2 k} \exp \left[-\frac{1}{2} \sum_\lambda a_\lambda y_\lambda^2 \right]. \quad (2.109)$$

The sum in this integrand can be reduced by considering the integral

$$\int_{-\infty}^{\infty} dy_i \ y_i y_j \left[-\frac{1}{2} a_i y_i^2 \right] = \sqrt{\frac{2\pi}{a_i^3}} \delta_{ij} \quad (2.110)$$

i.e. a standard Gaussian integral with the addition of a Kronecker delta. This is due to the fact that if $i \neq j$ then the integration is over an odd function which goes identically to zero. Otherwise it is an integration of the form $\int dy \ y^2 e^{-\frac{1}{2} a y^2}$. From this the correlation function can be evaluated

$$\langle x_j x_k \rangle = C \sum_{\lambda_1} e_{\lambda_1 j} e_{\lambda_1 k} \sqrt{\frac{2\pi}{a_{\lambda_1}^3}} \prod_{\lambda \neq \lambda_1} \sqrt{\frac{2\pi}{a_{\lambda}}}. \quad (2.111)$$

The normalisation constant C may also be expressed as a product of eigenvalues $C = \prod_{\lambda} \sqrt{\frac{a_{\lambda}}{2\pi}}$, and inserting this into the equation for the correlation function produces

$$\langle x_j x_k \rangle = \sum_{\lambda_1} \frac{1}{a_{\lambda_1}} e_{\lambda_1 j} e_{\lambda_1 k} = (A^{-1})_{jk}. \quad (2.112)$$

A Gaussian process is now obtainable from this distribution by attaching a time label to each random variable and taking the continuous limit. Before doing so, we shall consider the generalisation of this distribution to complex variables.

2.2.2 Complex Gaussian Distributions

The Gaussian process examined above uses real variables z_i , but later results require a generalisation to complex variables. Consider a zero-mean Gaussian distribution over the complex variables⁸ η, η^* defined by

$$W[\eta, \eta^*] = C \exp \left[-\frac{1}{2} \mathbf{z}^T \Phi \mathbf{z} \right] \quad (2.113)$$

⁸The complex process has to use both the variable and its conjugate to ensure that the integral over the distribution is both real valued and convergent.

$$\mathbf{z} = \begin{pmatrix} \eta \\ \eta^* \end{pmatrix} \quad \Phi = \begin{pmatrix} \phi_{\eta\eta} & \phi_{\eta\eta^*} \\ \phi_{\eta^*\eta} & \phi_{\eta^*\eta^*} \end{pmatrix}. \quad (2.114)$$

We can assume without loss of generality that the covariance matrix is real and symmetric, as it is always possible to redefine one's stochastic variables to satisfy this condition. The exponent can then be expressed in terms of two real variables:

$$\eta = x_1 + ix_2 \quad (2.115)$$

$$\eta^* = x_1 - ix_2 \quad (2.116)$$

which allows for a redefinition of the distribution exponent

$$\mathbf{z}^T \Phi \mathbf{z} = \mathbf{x}^T \Sigma \mathbf{x} \quad (2.117)$$

$$\mathbf{x} = \begin{pmatrix} x_1 \\ x_2 \end{pmatrix} \quad \Sigma = \begin{pmatrix} \sigma_{11} & \sigma_{12} \\ \sigma_{21} & \sigma_{22} \end{pmatrix}. \quad (2.118)$$

The elements of Σ are related to Φ in the following manner

$$\sigma_{11} = \phi_{\eta\eta} + 2\phi_{\eta\eta^*} + \phi_{\eta^*\eta^*} \quad (2.119)$$

$$\sigma_{22} = 2\phi_{\eta\eta^*} - \phi_{\eta\eta} - \phi_{\eta^*\eta^*} \quad (2.120)$$

$$\sigma_{12} = \sigma_{21} = i(\phi_{\eta\eta} - \phi_{\eta^*\eta^*}) \quad (2.121)$$

with a new determinant

$$|\Sigma| = -4(\phi_{\eta\eta}\phi_{\eta^*\eta^*} - \phi_{\eta\eta^*}^2). \quad (2.122)$$

Importantly, the new matrix Σ is also positive semi-definite.

To find the constant C integrate over the two complex variables:

$$\int d\eta d\eta^* C \exp \left[-\frac{1}{2} \mathbf{z}^T \Phi \mathbf{z} \right] = \int dx_1 dx_2 JC \exp \left[-\frac{1}{2} \mathbf{x}^T \Sigma \mathbf{x} \right] \quad (2.123)$$

$$J = \left| \frac{\partial z_i}{\partial x_j} \right| = \begin{vmatrix} 1 & i \\ 1 & -i \end{vmatrix} = -2i \quad (2.124)$$

where the usual identities for real Gaussian distributions can be employed

$$\int dx_1 dx_2 JC \exp \left[-\frac{1}{2} \mathbf{x}^T \Sigma \mathbf{x} \right] = -4\pi i C |\Sigma|^{-1/2} = 2\pi \sqrt{\frac{1}{\phi_{\eta\eta}\phi_{\eta^*\eta^*} - \phi_{\eta\eta^*}^2}} C \quad (2.125)$$

$$\implies C = \frac{\sqrt{\phi_{\eta\eta}\phi_{\eta^*\eta^*} - \phi_{\eta\eta^*}^2}}{2\pi} = \frac{|\Phi|}{2\pi}. \quad (2.126)$$

This is the same result as if we had used the identities for real Gaussian processes directly on Eq. (2.113). Correlation functions for these complex variables can similarly be calculated by transforming to the real variables. Using the inverse matrices

$$\Sigma^{-1} = \frac{1}{|\Sigma|} \begin{pmatrix} \sigma_{22} & -\sigma_{21} \\ -\sigma_{12} & \sigma_{11} \end{pmatrix} \quad (2.127)$$

$$\Phi^{-1} = \frac{1}{|\Phi|} \begin{pmatrix} \phi_{\eta^*\eta^*} & -\phi_{\eta^*\eta} \\ -\phi_{\eta\eta^*} & \phi_{\eta\eta} \end{pmatrix} \quad (2.128)$$

and the identity

$$\int dx_1 dx_2 \mathbf{x} \mathbf{x}^T JC \exp \left[-\frac{1}{2} \mathbf{x}^T \Sigma \mathbf{x} \right] = \Sigma^{-1} \quad (2.129)$$

then the correlation functions may be calculated:

$$\langle \eta \eta \rangle = \int d\eta d\eta^* \eta \eta C \exp \left[-\frac{1}{2} \mathbf{z}^T \Phi \mathbf{z} \right] = \frac{\phi_{\eta^* \eta}}{|\Phi|} = (\Phi^{-1})_{11} \quad (2.130)$$

$$\langle \eta^* \eta^* \rangle = \int d\eta d\eta^* \eta \eta C \exp \left[-\frac{1}{2} \mathbf{z}^T \Phi \mathbf{z} \right] = \frac{\phi_{\eta \eta}}{|\Phi|} = (\Phi^{-1})_{22} \quad (2.131)$$

$$\langle \eta \eta^* \rangle = \int d\eta d\eta^* \eta \eta C \exp \left[-\frac{1}{2} \mathbf{z}^T \Phi \mathbf{z} \right] = \frac{\phi_{\eta^* \eta}}{|\Phi|} = (\Phi^{-1})_{12}. \quad (2.132)$$

Thus, the complex Gaussian distribution in one variable obeys the same identities as in the two-variable real case. In fact, if we repeat the above process for complex entries in the covariance matrix, we only require that the matrix be positive semi-definite and Hermitian.

2.2.3 The Multivariate Complex Gaussian

We will now consider a distribution over N complex variables. The Gaussian distribution is now:

$$W[\eta_1, \eta_1^* \dots \eta_N, \eta_N^*] = \int d\eta_1 d\eta_1^* \dots d\eta_N d\eta_N^* C' \exp \left[-\frac{1}{2} \sum_{ij} \mathbf{z}^T \Phi \mathbf{z} \right]. \quad (2.133)$$

with complex variables are ordered in the following manner

$$\mathbf{z} = \begin{pmatrix} \mathbf{z}_1 \\ \mathbf{z}_2 \\ \vdots \\ \mathbf{z}_N \end{pmatrix} \quad \mathbf{z}_i = \begin{pmatrix} \eta_i \\ \eta_i^* \end{pmatrix}. \quad (2.134)$$

We begin by finding the new normalisation constant C' :

$$\int d\eta_1 d\eta_1^* \dots W[\eta_1, \eta_1^* \dots \eta_N, \eta_N^*] = 1 \quad (2.135)$$

The first step to performing this integral is decomposing the distribution exponent,

$$\sum_{ij} \mathbf{z}_i^T \Phi \mathbf{z}_j = \sum_{ij} \mathbf{z}_i^T \Phi_{ij} \mathbf{z}_j = \sum_{ij\alpha\beta} z_i^\alpha \Phi_{ij}^{\alpha\beta} z_j^\beta \quad (2.136)$$

where the extra indices act as the most convenient way to distinguish between each complex variable z_i^1 and its complex conjugate z_i^2 :

$$z_i^1 = \eta_i \quad z_i^2 = \eta_i^* \quad (2.137)$$

Since we already know the results for the single variable complex case, the easiest option here is to perform a spectral decomposition of the covariance matrix:

$$\Phi = \sum_{\lambda} a_{\lambda} \mathbf{e}_{\lambda} \mathbf{e}_{\lambda}^T \quad (2.138)$$

$$\Phi_{ij}^{\alpha\beta} = \sum_{\lambda} a_{\lambda} e_{i\lambda}^{\alpha} e_{j\lambda}^{\beta} \quad (2.139)$$

$$\sum_{ij} \mathbf{z}_i^T \Phi_{ij} \mathbf{z}_k = \sum_{ij\alpha\beta\lambda} a_{\lambda} z_i^{\alpha} e_{i\lambda}^{\alpha} e_{j\lambda}^{\beta} z_j^{\beta} = \sum_{\lambda} a_{\lambda} g_{\lambda}^2 \quad (2.140)$$

where in the final identity we have made use of

$$g_{\lambda} = \sum_{i\alpha} z_i^{\alpha} e_{i\lambda}^{\alpha}. \quad (2.141)$$

With this transformation, the integral over the distribution is

$$\int d\eta_1 d\eta_1^* \dots C' \exp \left[-\frac{1}{2} \sum_{ij} \mathbf{z}_i^T \Phi_{ij} \mathbf{z}_j \right] = \int dg_1 dg_2 \dots J C' \exp \left[-\frac{1}{2} \sum_{\lambda} a_{\lambda} g_{\lambda}^2 \right] \quad (2.142)$$

$$J = \left| \frac{\partial z_i^\alpha}{\partial g_\lambda} \right| = |e_{\lambda i}^\alpha| = 1. \quad (2.143)$$

Using the usual rules for Gaussian integrals this gives:

$$C' = (2\pi)^{N/2} \prod_\lambda \sqrt{\frac{1}{a_\lambda}} = \sqrt{\frac{(2\pi)^N}{|\Phi|}} \quad (2.144)$$

as expected. Correlation functions may be calculated in a similar manner:

$$\langle z_i^\alpha z_j^\beta \rangle = \sum_{\lambda\lambda'} e_{i\lambda}^\alpha e_{j\lambda'}^\beta \langle g_\lambda g_{\lambda'} \rangle \quad (2.145)$$

$$\langle g_\lambda g_{\lambda'} \rangle = \frac{1}{a_\lambda} \delta_{\lambda\lambda'} \quad (2.146)$$

$$\Rightarrow \langle z_i^\alpha z_j^\beta \rangle = \sum_\lambda \frac{1}{a_\lambda} e_{i\lambda}^\alpha e_{j\lambda}^\beta = \left(\Phi_{ij}^{\alpha\beta} \right)^{-1} \quad (2.147)$$

which encapsulates all of the correlation functions in terms of η_i and its conjugate

$$\langle \eta_i \eta_j^* \rangle = \langle z_i^1 z_j^2 \rangle = \left(\Phi_{ij}^{12} \right)^{-1} \quad (2.148)$$

$$\langle \eta_i \eta_j \rangle = \langle z_i^1 z_j^1 \rangle = \left(\Phi_{ij}^{11} \right)^{-1} \quad (2.149)$$

$$\langle \eta_i^* \eta_j^* \rangle = \langle z_i^2 z_j^2 \rangle = \left(\Phi_{ij}^{22} \right)^{-1} \quad (2.150)$$

.

The N dimensional complex Gaussian and the correlation functions generated from it have the same structure as in the case of a $2N$ dimensional real Gaussian. From this point forward we may apply these identities to any complex process.

2.2.4 The Hubbard-Stratonovich Transformation

Having established the important properties of the complex Gaussian process, we will examine the effect of Fourier transforming the probability distribution to the z_i conjugate variable k_i :

$$\begin{aligned}\kappa(k_1 \dots k_N) &= \int d\mathbf{z} \, W(\mathbf{z}) e^{i\mathbf{z}^T \mathbf{k}} \\ &= \int d\mathbf{z} \, \exp \left[-\frac{1}{2} \mathbf{z}^T \Phi \mathbf{z} + i\mathbf{z}^T \mathbf{k} \right].\end{aligned}\quad (2.151)$$

Here \mathbf{z} is the vector of all the complex variables and its conjugates, with individual elements labeled as before with z_i^α . Evaluating the Fourier transform is simply a case of completing the square of the exponent:

$$-\frac{1}{2} \mathbf{z}^T \Phi \mathbf{z} + i\mathbf{z}^T \mathbf{k} = -\frac{1}{2} (\mathbf{z} - i\Phi^{-1}\mathbf{k})^T \Phi (\mathbf{z} - i\Phi^{-1}\mathbf{k}) - \frac{1}{2} \mathbf{k}^T \Phi^{-1} \mathbf{k}. \quad (2.152)$$

Since the integration over \mathbf{z} is shifted only by a constant $-i\Phi^{-1}\mathbf{k}$, the Jacobian of this translation is 1. Therefore the Fourier transform of the probability distribution is simply

$$\kappa[\mathbf{k}] = e^{-\frac{1}{2} \mathbf{k}^T \Phi^{-1} \mathbf{k}}. \quad (2.153)$$

The exponent of this expression may be expanded in terms of the random variable correlations:

$$\mathbf{k}^T \Phi^{-1} \mathbf{k} = \sum_{ij\alpha\beta} k_i^\alpha \langle z_i^\alpha z_j^\beta \rangle k_j^\beta \quad (2.154)$$

As has already been mentioned, each random variable has an ordering parameter

t_i . Much like in the path integral derivation, we can make these ordering parameters evenly spaced by a gap Δ . Then if $t_N = t_f$, $t_0 = 0$, the gap is given by $\Delta = \frac{t_f}{N}$. Defining now a single process $z_i^\alpha = z^\alpha(t_i)$ $k_i^\alpha = \Delta k^\alpha(t_i)$, we take the continuum limit $N \rightarrow \infty$. In this limit, vector and matrix products become integrals

$$\sum_i z_i^\alpha k_i^\alpha = \sum_i \Delta z^\alpha(t_i) k^\alpha(t_i) \rightarrow \int_0^{t_f} dt z^\alpha(t) k^\alpha(t) \quad (2.155)$$

$$\sum_{ij} k_i^\alpha \left\langle z_i^\alpha z_j^\beta \right\rangle k_j^\beta = \sum_{i,j} \Delta^2 k^\alpha(t_i) A^{\alpha\beta}(t_i, t_j) k^\beta(t_j) \rightarrow \int_0^{t_f} \int_0^{t_f} dt dt' k^\alpha(t) A^{\alpha\beta}(t, t') k^\beta(t'). \quad (2.156)$$

Here the matrix $A^{\alpha\beta}$ is defined in relation to $\Phi^{\alpha\beta}$ in the following manner:

$$\sum_\beta \int_0^{t_f} dt' \Phi^{\alpha\beta}(t, t') A^{\beta\gamma}(t', t'') = \delta(t - t'') \delta_{\alpha\gamma} \quad (2.157)$$

$$A^{\alpha\beta}(t, t') = \left\langle z_i^\alpha(t) z_j^\beta(t') \right\rangle_{\mathbf{z}}. \quad (2.158)$$

Having taken the continuous limit, the measure for the integration is now akin to a path integral, as $\lim_{N \rightarrow \infty} \prod_i dz_i^\alpha \rightarrow \mathcal{D}z^\alpha(\tau)$. In the continuous limit, the Fourier transform κ becomes

$$\kappa(\mathbf{k}(t_f)) = \exp \left[-\frac{1}{2} \sum_{\alpha\beta} \int_0^{t_f} \int_0^{t_f} dt dt' k^\alpha(t) A^{\alpha\beta}(t, t') k^\beta(t') \right]. \quad (2.159)$$

Remembering the original definition of κ in Eq. (2.151), it is possible to interpret this not just as a Fourier transform but as a functional average

$$\kappa(\mathbf{k}(t_f)) = \left\langle \exp \left[i \sum_\alpha \int_0^{t_f} dt z^\alpha(t) k^\alpha(t) \right] \right\rangle_{\mathbf{z}}. \quad (2.160)$$

Importantly, the relationship between the k^α is *not* constrained in the same way as the

variables z^α are. This means we are free to choose what, if any, functional dependence there is between $k^1(t)$ and $k^2(t)$.

Putting all of this together gives us the HS transformation⁹:

$$\left\langle \exp \left[i \sum_{\alpha} \int_0^{t_f} dt \ z^{\alpha}(t) k^{\alpha}(t) \right] \right\rangle_{\mathbf{z}} \quad (2.161)$$

$$= \exp \left[-\frac{1}{2} \sum_{\alpha\beta} \int_0^{t_f} \int_0^{t_f} dt dt' \ k^{\alpha}(t) \langle z^{\alpha}(t) z^{\beta}(t') \rangle k^{\beta}(t') \right] \quad (2.162)$$

which is easily generalised to multivariate processes

$$\left\langle \exp \left[i \sum_{i\alpha} \int_0^{t_f} dt \ z_i^{\alpha}(t) k_i^{\alpha}(t) \right] \right\rangle_{\mathbf{z}} \quad (2.163)$$

$$= \exp \left[-\frac{1}{2} \sum_{ij\alpha\beta} \int_0^{t_f} dt \int_0^{t_f} dt' \ k_i^{\alpha}(t) \langle z_i^{\alpha}(t) z_j^{\beta}(t') \rangle k_j^{\beta}(t') \right]. \quad (2.164)$$

The HS transformation equates a deterministic non-local integral exponent to one involving local stochastic terms that must be averaged over the distribution W . In a more physical sense, we can consider the HS transformation as converting a system of two body potentials into a set of independent particles in a fluctuating field. It is a tremendously useful transformation, and in the next chapter shall be applied in concert with the influence functional formalism.

2.2.5 Hubbard-Stratonovich transformation over P time dimensions

In order to apply the HS transformation to later results, we provide here an original generalisation of the transformation. In the previous sections, the introduction of

⁹Invented by Stratonovich, popularised outside the USSR by Hubbard.

extra stochastic variables and their time dynamics was achieved simply through the addition of extra indices, partitioning the arbitrary sum of random complex variables in different ways. There is no reason the same technique cannot be recycled to introduce additional time dimensions. Consider the Fourier transform in Eq. (2.151), partitioning \mathbf{z} into J variables of length N . This time index can be further divided into P pieces of length M (i.e. $N = PM$)

$$\mathbf{z}^T \mathbf{k} = \sum_{\alpha=1}^2 \sum_{j=1}^J \sum_{n=1}^N z_{nj}^{\alpha} k_{nj}^{\alpha} = \sum_{\alpha=1}^2 \sum_{j=1}^J \sum_{p=1}^P \sum_{m=1}^M z^{\alpha}(t_m^p) k^{\alpha}(t_m^p) \quad (2.165)$$

$$z_{nj}^{\alpha} k_{nj}^{\alpha} = z_j^{\alpha}(t_m^p) k_j^{\alpha}(t_m^p) \quad p = \left\lfloor \frac{n}{M} \right\rfloor \quad m = n \bmod (M). \quad (2.166)$$

If we take the continuum limit $M, P \rightarrow \infty$ and assign $t_1^p = 0$, $t_M^p = t_f^p$, then the average over $e^{i\mathbf{z}^T \mathbf{k}}$ can be evaluated in the same way as Eq. (2.151):

$$\begin{aligned} & \left\langle \exp \left[i \sum_{\alpha i p} \left(\int_0^{t_f^p} dt^p z_i^{\alpha}(t^p) k_i^{\alpha}(t^p) \right) \right] \right\rangle_{\mathbf{z}} \\ &= \exp \left[-\frac{1}{2} \sum_{ij\alpha\beta p\bar{p}} \int_0^{t_f^p} \int_0^{t_f^{\bar{p}}} dt^p dt^{\bar{p}} k_i^{\alpha}(t^p) A_{ij}^{\alpha\beta}(t^p, t^{\bar{p}}) k_j^{\beta}(t^{\bar{p}}) \right] \end{aligned} \quad (2.167)$$

$$A_{ij}^{\alpha\beta}(t^p, t^{\bar{p}}) = \left\langle z_i^{\alpha}(t^p) z_j^{\beta}(t^{\bar{p}}) \right\rangle_{\mathbf{z}} \quad (2.168)$$

This is the HS transformation in P time dimensions. This equation is a crucial generalisation required to move beyond the partitioned approximation when evaluating influence functionals, whose full utility will become apparent in the next chapter.

2.3 Koopman von-Neumann Dynamics

We close this chapter introducing the Koopman von-Neumann (KvN) formalism for classical mechanics. This is in a sense the adjoint to formulations of quantum mechanics in phase space [112] or path integrals. While the latter theories are “classicalised” descriptions of quantum phenomena, KvN mechanics casts classical systems in a quantum language. Specifically, KvN mechanics is the reformulation of classical mechanics in a Hilbert space formalism [113]. This representation [113] has been deployed fruitfully in various contexts, including the time-dependent frequency harmonic oscillator [114], the analysis of dissipative phenomena [115], and in linear representations of non-linear dynamics [116]. In our context, KvN is a formalism that will allow for the direct importation of many of the quantum results of this thesis into a classical setting.

2.3.1 The Koopman Operator

KvN is a Hilbert space theory, so to begin with let us define its essential characteristics. In its functional form, a Hilbert space consists of the set of functions $L^2(\mathcal{P}, d\mu)$:

$$L^2(\mathcal{P}, d\mu) = \left\{ \phi : \mathcal{P} \rightarrow \mathbb{C} \left| \int d\mu |\phi|^2 < \infty \right. \right\} \quad (2.169)$$

i.e. the set of all functions on a space \mathcal{P} that are square integrable with a measure $d\mu$. The other necessary ingredient in a Hilbert space is the definition of an inner product

$$\langle \phi | \psi \rangle = \int d\mu \phi^* \psi \quad (2.170)$$

for $\phi, \psi \in L^2(\mathcal{P}, d\mu)$.

Physics is introduced to this formalism by interpreting the elements of L^2 as probability density amplitudes. In addition, observables are associated with Hermitian

operators and states obey the Born rule. The probability distribution for a state is therefore the square of its wavefunction $\rho = |\psi|^2$. In addition, Stone's theorem guarantees there exists a one-parameter, continuous group of unitary transformations on L^2 of the form [117]:

$$\hat{U}(t) = e^{i\hat{A}t} \quad (2.171)$$

where \hat{A} is a unique *self-adjoint* operator¹⁰. This family of transformations is interpreted as time evolution and leads to the following differential equation:

$$\dot{\psi} = i\hat{A}\psi \quad (2.172)$$

So far, this is identical to quantum mechanics. The key distinction between quantum and Koopman dynamics is the way elements of L^2 are evolved in time. Specifying the form of \hat{A} adds physics to the formalism, and requires both the imposition of the Ehrenfest theorems, *and* a fundamental commutation relation [118]. The choice of commutation relation is the sole distinction between Koopman dynamics and quantum mechanics¹¹. Explicitly, the Ehrenfest theorems are

$$\frac{d\langle\hat{x}\rangle}{dt} = \left\langle \frac{\hat{p}}{m} \right\rangle \quad (2.173)$$

$$\frac{d\langle\hat{p}\rangle}{dt} = -\left\langle \hat{V}'(x) \right\rangle \quad (2.174)$$

since these hold for any state, we find the following relations for the time generator:

¹⁰This will be relevant in chapter 5.

¹¹It should also be noted that the choice of commutation relation also defines the measure on the Hilbert space. While $d\mu = dqdp$ is a valid measure in the classical case, only $d\mu = dq$ (q representation) preserves non-negativity for the quantum commutation relation [119]. One may still elect to use the full phase space quantum mechanically, but the object under consideration in this case is the Wigner quasiprobability distribution [120–122].

$$i \left[\hat{A}, \hat{x} \right] = \frac{\hat{p}}{m} \quad (2.175)$$

$$i \left[\hat{A}, \hat{p} \right] = -\hat{V}'(x) \quad (2.176)$$

In the quantum case $[\hat{x}, \hat{p}] = i\hbar$. When this is applied to Eqs. (2.175) & (2.176) they uniquely identify the self-adjoint operator¹² as $\hat{A} = -\frac{1}{\hbar}\hat{H}$. This recovers the familiar Schrödinger equation:

$$i\hbar\dot{\psi}_{\text{qm}} = \hat{H}\psi_{\text{qm}} \quad (2.177)$$

In KvN mechanics $[\hat{x}, \hat{p}] = 0$. As a result, the \hat{x} and \hat{p} operators have a common set of eigenstates. These form an orthonormal eigenbasis, furnished with the usual relationships:

$$\hat{x} |x, p\rangle = x |x, p\rangle \quad \langle x, p | x', p' \rangle = \delta(x - x') \delta(p - p')$$

$$\hat{p} |x, p\rangle = p |x, p\rangle \quad \int dx dp |x, p\rangle \langle x, p| = 1. \quad (2.178)$$

One consequence of allowing the phase space operator to commute is that it is *impossible* to construct an operator that satisfies the Ehrenfest theorems purely out of \hat{x} and \hat{p} . It is therefore necessary to introduce two new operators, $\hat{\lambda}$ and $\hat{\theta}$ with the commutation relations:

$$[\hat{x}, \hat{\lambda}] = [\hat{p}, \hat{\theta}] = i \quad (2.179)$$

$$[\hat{\lambda}, \hat{\theta}] = [\hat{\lambda}, \hat{p}] = [\hat{\theta}, \hat{x}] = 0 \quad (2.180)$$

With the addition of these “ambiguity” operators, one obtains $\hat{A} = -\hat{K}$, where \hat{K} is

¹²While it's easy to see that the first Ehrenfest theorem requires the time generator to consist of a $\frac{1}{2m}\hat{p}^2$ term, finding $\hat{V}(x)$ with the second Ehrenfest theorem necessitates the introduction of the overlap $\langle \hat{x} | \hat{p} \rangle = e^{\frac{ipx}{\hbar}}$ together with judicious use of integration by parts.

the *Koopman operator*:

$$\hat{U}_{\text{cl}}(t) = e^{-it\hat{K}} \quad (2.181)$$

$$\hat{K} = \frac{\hat{p}}{m} \hat{\lambda} - \hat{V}'(x) \hat{\theta} \quad (2.182)$$

2.3.2 Basis overlaps

In quantum mechanics, the position and momentum bases form a complementary pair, and it is often useful to transform between them. To do so, one must derive the *overlap* between them. This is particularly useful for specifying a representation of an operator in its conjugate basis. In KvN, basis transformations are similarly helpful. In the classical case there are four “canonical” sets of simultaneous eigenbases, these are:

$$\begin{aligned} |x, p\rangle \quad |x, \theta\rangle \\ |\lambda, p\rangle \quad |\lambda, \theta\rangle. \end{aligned} \quad (2.183)$$

Here we outline the procedure for deriving the overlap between two bases of non-commuting operators. Take two Hermitian operators \hat{x} and \hat{y} with the commutation relation:

$$[\hat{x}, \hat{y}] = 1 \quad (2.184)$$

it follows that

$$[\hat{x}, e^{a\hat{y}}] = ae^{a\hat{y}}. \quad (2.185)$$

Applying this commutator to an eigenstate of \hat{x} we obtain:

$$\hat{x}e^{a\hat{y}}|x\rangle = (x + a)e^{a\hat{y}}|x\rangle \quad (2.186)$$

indicating $e^{a\hat{y}}|x\rangle$ is an eigenstate of the \hat{x} operator with eigenvalue $x + a$. From this

we can conclude that $e^{a\hat{y}}$ is a *translation* operator¹³

$$e^{a\hat{y}} |x\rangle = |x + a\rangle. \quad (2.187)$$

Furthermore, it is possible give an explicit form for \hat{y} in this basis:

$$\langle x | \hat{y} | \psi \rangle = \lim_{a \rightarrow 0} \frac{1}{a} \langle x | e^{a\hat{y}} - 1 | \psi \rangle = \lim_{a \rightarrow 0} \frac{1}{a} (\psi(x + a) - \psi(x)) = \frac{\partial}{\partial x} \psi(x) \quad (2.188)$$

i.e. \hat{y} is given by $\frac{\partial}{\partial x}$ in the x representation.

In KvN mechanics, the commutator between operators is always i . Making the assignment $i\hat{y} = \hat{\lambda}$, we can calculate the overlap between \hat{x} and $\hat{\lambda}$:

$$\lambda \langle \lambda | x \rangle = \langle \lambda | i\hat{y} | x \rangle = i \frac{\partial}{\partial x} \langle \lambda | x \rangle \quad (2.189)$$

$$\implies \langle \lambda | x \rangle = N(\lambda) e^{-i\lambda x}. \quad (2.190)$$

The normalisation of the overlap is easily checked using:

$$\delta(x - x') = \int d\lambda \langle x' | \lambda \rangle \langle \lambda | x \rangle = 2\pi |N(\lambda)|^2 \delta(x - x') \quad (2.191)$$

$$\implies N(\lambda) = \frac{1}{\sqrt{2\pi}} \quad (2.192)$$

This generically specifies the form of the overlap between eigenstates. Any eigenbasis of an operator is also an eigenbasis of operators it commutes with. Equipped with this, one may straightforwardly generate the following overlaps for the simultaneous eigenstates

$$\langle x, \theta | x', p \rangle = \frac{1}{\sqrt{2\pi}} \delta(x - x') e^{-i\theta p} \quad (2.193)$$

¹³Strictly speaking we only have $e^{a\hat{y}} |x\rangle = f(x, y) |x + a\rangle$, but normalisation of eigenstates guarantees $f(x, y)$ is a pure phase. We can then get rid of this phase with a redefinition of the instantaneous eigenstates. All I'm doing here is skipping the baggage irrelevant to the final result.

$$\langle \lambda, p | x, p' \rangle = \frac{1}{\sqrt{2\pi}} \delta(p - p') e^{-i\lambda x} \quad (2.194)$$

$$\langle \lambda, p | x, \theta \rangle = \int dx' dp' \langle \lambda, p | x', p' \rangle \langle x', p' | x, \theta \rangle = \frac{1}{2\pi} e^{-i\lambda x} e^{i\theta p}. \quad (2.195)$$

The mathematics of specifying overlaps is generic between quantum and KvN mechanics, with the only generalisation arising from KvN's simultaneous eigenbases allowing a greater degree of freedom in representation.

2.3.3 Liouville's Theorem For KvN

Having derived the Koopman operator, we should check that this is consistent with more standard formulations of classical dynamics. Taking the evolution equation

$$i \frac{d}{dt} |\psi\rangle = -\hat{K} |\psi\rangle \quad (2.196)$$

we pick a specific representation:

$$\psi(x, p) = \langle x, p | \psi \rangle \quad (2.197)$$

$$\hat{x} \rightarrow x \quad \hat{p} \rightarrow p \quad (2.198)$$

$$\hat{\lambda} \rightarrow -i \frac{\partial}{\partial x} \quad \hat{\theta} \rightarrow -i \frac{\partial}{\partial p} \quad (2.199)$$

which leads to

$$i\dot{\psi} = \frac{p}{m} \frac{\partial \psi}{\partial x} - V'(x) \frac{\partial \psi}{\partial p}. \quad (2.200)$$

This evolution equation may be expressed via the Poisson bracket

$$\{H, \odot\} = \frac{\partial H}{\partial x} \frac{\partial \odot}{\partial p} - \frac{\partial H}{\partial p} \frac{\partial \odot}{\partial x}. \quad (2.201)$$

The evolution of a classical wavefunction may therefore be expressed as

$$\dot{\psi}(q, p) = i\hat{K}\psi(q, p) = \{H, \psi(q, p)\} \quad (2.202)$$

The phase space representation of the Koopman operator is the *Poisson bracket*. The evolution equation for the classical wavefunction is therefore identical to that for the associated probability density

$$\dot{\rho}(q, p) = \{H, \rho(q, p)\}. \quad (2.203)$$

This fact is particularly helpful, as it means that a classical wavefunction and its equivalent probability density are evolved by the *same* propagator. Explicitly:

$$\hat{U}_{\text{cl}}(t) = e^{-it\hat{K}} \quad (2.204)$$

$$\psi(x_f, p_f) = U_{\text{cl}}(x_f, p_f, t_f; x_i, p_i, 0) \psi(x_i, p_i) \quad (2.205)$$

$$|\psi(x_f, p_f)|^2 = \rho(x_f, p_f) = U_{\text{cl}}(x_f, p_f, t_f; x_i, p_i, 0) \rho(x_i, p_i). \quad (2.206)$$

This identification of the Koopman operator with the Poisson bracket is doubly useful, as any coordinate transformation that preserves the Poisson bracket (i.e. canonical transformations) [1] can be used as an operator basis. This is an important distinction from quantum mechanics where the quantisation of a Hamiltonian must be done in the Cartesian coordinate system. This is due to the fact that different physical systems can be represented by the same mathematical expression. Consider that $H = \frac{p^2}{2}$ can equally well represent a free particle or harmonic oscillator with a set of canonical coordinates. Classically, this is irrelevant as one can freely transform the dynamics from one set of coordinates to another. When phase space measurements do not commute however, the statistics of a free particle and harmonic oscillator can *not* be brought together with a canonical coordinate transformation. In this sense

the fundamental commutation relation forces a particular coordinate basis on the operators we quantise, whereas in KvN we are free to transform operators with any transformation that preserves the Poisson bracket¹⁴.

Aside: Unifying Classical and Quantum Dynamics

One of the most intriguing aspects of KvN is the prospect of a truly unified formalism for both quantum and classical mechanics. This was achieved recently in a framework known as Operant Dynamic Conditioning (ODC) [105, 118]. Briefly, this is accomplished by extending the usual quantum mechanical algebra with ambiguity operators:

$$[\hat{x}_q, \hat{p}_q] = i\kappa\hbar \quad [\hat{x}_q, \hat{\lambda}_q] = [\hat{p}_q, \hat{\theta}_q] = i \quad (2.207)$$

where κ is a parameter one sets between the *limit* of 0 (classical) and 1 (quantum). This is precisely isomorphic to the KvN algebra, such that the quantum operators may all be expressed as linear combinations of *classical* KvN operators. Imposing the condition that in the limit $\kappa \rightarrow 0$ the quantum operators become their classical equivalents, it is possible to derive the generator of both quantum and classical mechanics in Eq. (2.171) as:

$$\frac{1}{\hbar}\hat{\mathcal{H}}_{qc} = \frac{1}{m}\hat{p}\hat{\lambda} + \frac{1}{\hbar\kappa} \left[V\left(\hat{x} - \frac{\hbar\kappa}{2}\hat{\theta}\right) - V\left(\hat{x} + \frac{\hbar\kappa}{2}\hat{\theta}\right) \right] \quad (2.208)$$

The generator bears more than a passing resemblance to the Wigner phase function [123], which should not be surprising. The Wigner phase represents quantum mechanics in a classical phase space, while the ODC generator determines time evo-

¹⁴One might ask about what becomes of the commutation relations between the phase space variables and the ambiguity variables after a coordinate transform. Given that these operators are invoked precisely to enforce the Ehrenfest theorem (and therefore the Poisson bracket), the commutation relationships between the transformed ambiguity and phase space operators are identical to the original coordinates.

lution purely in terms of the classical operators. This generator also transparently recovers the Koopman operator in the limit¹⁵

$$\lim_{\kappa \rightarrow 0} \hat{\mathcal{H}}_{\text{qc}} = \hbar \hat{K}. \quad (2.209)$$

There is a supreme irony present in this combined picture of dynamics, but it requires some context. Since the genesis of quantum mechanics, there has always been an argument over how to interpret the formalism, and the unintuitive behaviour it predicts. This debate has spawned plenty of alternate interpretations [125], most of which have the questionable virtue of being wildly unparsimonious without achieving a more intelligible rendering of the facts of reality. One of the earliest proposals to “explain” quantum mechanics was that as a theory it was incomplete, and as such its predictions are due to some missing information¹⁶.

Specifically, it was proposed that *local* hidden variables were a necessary component of any theory [126] that could surmount the EPR paradox [127]. Implicit in this work is an expectation that by extending quantum theory, one could restore counterfactual definiteness to the situation and reclassicalise the world. This was a forlorn hope, as later work by Bell demonstrated that no local hidden variable theory could capture the correlative predictions of quantum mechanics [128], subsequently confirmed by the experiments of Alain Aspect [129]. As with any no-go theorem there are a number of loopholes, but subsequent [130] experiments [131] have [132] closed [133] these [134] off [135].

¹⁵Astonishingly, quadratic polynomial potentials automatically satisfy $\hat{\mathcal{H}}_{\text{qc}} = \hbar \hat{K}$ for any value of κ . This is emphasising the same point as the semiclassical approximation for the path integral, namely that for these potentials the quantum and classical dynamics are *identical*. The principal distinction between the regimes is that when κ is non-zero, the xp representation of the wavefunction is essentially the Wigner quasiprobability. Its negative regions are of the order $\hbar\kappa$, so that in the classical limit we obtain a regular probability density [124].

¹⁶A view favoured by both Einstein & de Broglie. In their attempts to pursue it, Einstein kept deriving self-evidently absurd results, while de Broglie formulated the pilot wave theory that became de Broglie-Bohm mechanics.

The consequence of this work is the confirmation that it is impossible to describe quantum mechanics in a classical setting with local hidden variables. No great surprise, but KvN dynamics provides a complement to this statement- it *is* possible to describe classical physics in a quantum mechanical framework, but requires *local hidden variables*¹⁷.

Freeman Dyson once remarked that it may well be that the book of the world is written in a language humans are incapable of understanding. Nevertheless, the universe exists and has structure, so the minimum we can expect is structure in its statistics. In this view, it seems likely that our most fundamental understanding of reality has to be on a statistical level. If it happens to be possible to describe the dynamics of those statistics, so much the better. ODC is just such a statistical theory. Its principal advantage is the unity of description that it provides, manifestly containing classical physics as a special case, but *retaining* the essential novelty of quantum mechanics (i.e. statistics on a non-commutative algebra).

Finally, let me remark that there is an ongoing debate in foundational quantum mechanics as to the epistemic or ontic nature¹⁸ of the wavefunction [136], but I have never met a physicist similarly concerned about the nature of a probability distribution. This seems to be a case of cognitive dissonance, as the two are in a profound sense the same object. It is perhaps a historical misfortune that almost all human experience happens at the pathological end of the scale where $\kappa = 0$ and all the distributions are delta functions. The fact that this particular regime is familiar tricks us into believing we understand it better than we do.

I would hazard that the vague sense of discomfort experienced by those learning

¹⁷One could reframe this and say that quantum mechanics *is* a local hidden variable theory, but so is classical mechanics, and this doesn't remove the essential "problem" of non-commuting variables. This rather defeats the point of invoking them in a quantum context.

¹⁸Stephen Weinberg has a nice line on the encroachment of philosophical vocabulary into physics: "A physicist friend of mine once said that in facing death he drew some consolation from the reflection that he would never again have to look up the word hermeneutics in the dictionary."

quantum mechanics for the first time is misplaced. The ultimate cause is that classical mechanics is the strange theory, representing a limit which breaks algebraic structure. The fact the two theories are difficult to reconcile is not due to inherent quantum spookiness, but an under-interpretation of classical physics.

2.4 Chapter Summary

In this chapter we have presented the essential techniques necessary for the derivations in the rest of the thesis. Many of these methods have a rich literature surrounding them, and in a more extended analysis each could warrant a chapter of their own. In the process I have attempted to provide where possible some novel derivations for important results¹⁹. I have also included one original result in this chapter- the P time HS transform seen in section 2.2.5, on the grounds that it is easier to read about the generalisation when it is immediately preceded by the original transformation. This will be utilised in chapter 3, together with the influence functional formalism.

The Koopman von-Neumann formalism is exclusively used in chapter 5, where we will derive its path integral before bringing to bear all the methods detailed in this chapter to perform an equivalent classical analysis to that found in chapter 3.

¹⁹It is surprisingly difficult to source the derivations for some of these results. For example, I was unable to find a complete calculation for the forced harmonic oscillator path integral before embarking on my own.

Chapter 3

The Extended Stochastic Liouville Equation

Man is sometimes extraordinarily,
passionately, in love with suffering.

Fyodor Dostoevsky

Equipped with the mathematical techniques detailed in the previous chapter, we may turn our attention to deriving the central result of this thesis. This chapter adapts and extends my previously published work [137], deriving using the path integral formalism a set of stochastic differential equations for the reduced density matrix of an open system which describe its dynamics *exactly*. The model employed is based on a generalised Caldeira-Leggett Hamiltonian [90], from which we obtain a system of first order stochastic differential equations over real and imaginary time that after averaging describe the evolution of the state of a dissipative quantum system for partition-free initial conditions. These equations, which we term the Extended Stochastic Liouville Equation (ESLE), represent both a synthesis and extension of the work outlined in the introduction, allowing for a simple and exact closed form description of an arbitrary open system evolving from realistic initial conditions. The

chapter will be organised as follows:

Section 3.1 details the model employed, and the class of applicable initial conditions. In section 3.2 we will employ the influence functional formalism (sec. 2.1.8) to explicitly derive the effective propagator for the open system. In section 3.3 the two-time HS transformation (sec. 2.2.5) is applied to the influence functional found in the previous section, introducing the corresponding complex Gaussian stochastic fields. Section 3.4 presents the path integral describing the reduced density matrix of the primary system and the operator ESLE equations of motion that it implies. These equations account for both the generalised Hamiltonian and partition-free initial conditions.

After deriving the ESLE, we present some brief extensions in section 3.5. These include a model simplification for an infinite bath, a classical limit for the ESLE, an approximation for the Hamiltonian of mean force, and an examination of the effect of attaching a second environment. Finally, section 3.6 provides a short summary and discussion of the ESLE.

3.1 Model

Consider a many-body phonon system of the type described in section 1.3. It consists of a general central system (the open system), described by the Hamiltonian $H_Q(q)$. The secondary system (the environment) is composed of M harmonic oscillators (with masses m_i) coupled both internally and with the open system. The open system itself may be subjected to time-dependent external fields. The environment uses displacement coordinates ξ_i and the interaction between the two systems is linear in $\xi \equiv \{\xi_i\}$ but a set of arbitrary functions $f \equiv \{f_i\}$ in q . The joint Hamiltonian is therefore

$$H_{\text{tot}}(q, \xi) = H_Q(q) + \frac{1}{2} \sum_{i=1}^M m_i \dot{\xi}_i^2 + \frac{1}{2} \sum_{i,j=1}^M A_{ij} \xi_i \xi_j - \sum_i^M f_i(q) \xi_i. \quad (3.1)$$

This Hamiltonian differs from the standard CL Hamiltonian in Eq. (1.22) in two important respects. First, the interaction between the primary and secondary systems is no longer strictly bilinear, but may depend on any function of q . In addition, the atomic displacements that form the environment are now coupled to each other as well as the system, with the coupling described by the force-constant matrix A_{ij} . These alterations will have a material effect on the results. The counter-term found in Eq. (1.22) has been dropped, as this term disappears when the Hamiltonian of an arbitrary combined system is expanded in a power series in terms of atomic displacements. In this sense the model Hamiltonian is the second-order expansion of *any* conceivable system-bath Hamiltonian.

The density matrix evolves in the usual manner according to the Liouville equation:

$$\hat{\rho}^{\text{tot}}(t) = \hat{U}(t; t_0) \hat{\rho}^{\text{tot}}(t_0) \hat{U}^\dagger(t; t_0) \quad (3.2)$$

where

$$\hat{U}(t; t_0) = \hat{T} \exp \left[-\frac{i}{\hbar} \int_{t_0}^t dt' \hat{H}_{\text{tot}}(t') \right] \quad (3.3)$$

is the corresponding evolution operator (using the time ordering operator \hat{T}). Importantly we need not assume that the system Hamiltonian $H_Q(q)$ is time-independent, $H_Q(q) \equiv H_Q(q, t)$. The dynamics of the open system are found by tracing the full density matrix over the ξ coordinates:

$$\hat{\rho}(t) = \text{Tr}_\xi [\hat{\rho}^{\text{tot}}(t)] \quad (3.4)$$

while the total and reduced density matrices in coordinate space are, respectively:

$$\rho_t^{\text{tot}}(q, \xi; q', \xi') = \langle q, \xi | \hat{\rho}^{\text{tot}}(t) | q', \xi' \rangle \quad (3.5)$$

$$\rho_t(q, q') = \langle q | \hat{\rho}(t) | q' \rangle. \quad (3.6)$$

The propagators in this space are given by:

$$U(q, \xi, t; \bar{q}, \bar{\xi}, t_0) = \langle q, \xi | \hat{U}(t; t_0) | \bar{q}, \bar{\xi} \rangle \quad (3.7)$$

$$\langle \bar{q}, \bar{\xi} | \hat{U}^\dagger(t; t_0) | q, \xi \rangle = \langle \bar{q}, \bar{\xi} | \hat{U}(t_0; t) | q, \xi \rangle = U(\bar{q}, \bar{\xi}, t_0; q, \xi, t) \quad (3.8)$$

The second equality has been constructed to demonstrate that in coordinates, \hat{U}^\dagger has the form of a backward propagation in time. Setting $t_0 = 0$ for convenience, the open system density matrix in the coordinate representation is:

$$\rho_t(q, q') = \int d\bar{\xi} d\bar{\xi}' d\bar{q} d\bar{q}' d\xi d\xi' \delta(\xi - \xi') U(q, \xi, t; \bar{q}, \bar{\xi}, 0) \rho_0^{\text{tot}}(\bar{q}, \bar{\xi}; \bar{q}', \bar{\xi}') U(\bar{q}', \bar{\xi}', 0; q', \xi', t). \quad (3.9)$$

3.1.1 Normal Modes

To proceed we transform to a normal mode representation $\xi \rightarrow x = \{x_\lambda\}$, where

$$x_\lambda = \sum_i^M \sqrt{m_i} e_{\lambda i} \xi_i, \quad \xi_i = \sum_\lambda^M \frac{1}{\sqrt{m_i}} e_{i\lambda} x_\lambda$$

and $e_\lambda = \{e_{\lambda i}\}$ are eigenvectors of the dynamical matrix $D = \{D_{ij}\}$, $D_{ij} = \Lambda_{ij}/\sqrt{m_i m_j}$, with eigenvalues ω_λ^2 . The eigenvectors satisfy the usual orthogonality $e_\lambda^T e_{\lambda'} = \delta_{\lambda\lambda'}$, and completeness $\sum_\lambda e_\lambda e_\lambda^T = 1$ conditions. Applying these transformations, the Hamiltonian can be expressed as:

$$H_{\text{tot}}(q, x) = H_Q(q) + \frac{1}{2} \sum_{\lambda=1}^M (\dot{x}_\lambda^2 + \omega_\lambda^2 x_\lambda^2) - \sum_{\lambda} g_\lambda(q) x_\lambda \quad (3.10)$$

$$g_\lambda(q) = \sum_i^M \frac{1}{\sqrt{m_i}} e_{\lambda i} f_i(q), \quad f_i(q) = \sqrt{m_i} \sum_{\lambda} e_{i\lambda} g_\lambda(q). \quad (3.11)$$

The reduced density matrix is now:

$$\rho_t(q, q') = \int d\bar{x} d\bar{x}' dx d\bar{q} d\bar{q}' U(q, x, t; \bar{q}, \bar{x}, 0) \rho_0^{\text{tot}}(\bar{q}, \bar{x}; \bar{q}', \bar{x}') U(\bar{q}', \bar{x}', 0; q', x, t). \quad (3.12)$$

Before Eq. (3.12) can be solved, we must specify the form of the initial density matrix ρ_0^{tot} . It is usually assumed that the two systems are initially partitioned. The initial condition of Eq. (1.23) under strong coupling may not exactly predict the transient behaviour under perturbations due to the artificial equilibration of each system separately. If we wish to extract the exact transient dynamics of an open system (particularly at strong coupling) we must use a more realistic, non-partitioned initial condition. One solution employed by Grabert *et al.* [64] is to consider the full interacting system as being allowed to equilibrate with some time-independent Hamiltonian H_0 before applying any time-dependent perturbation:

$$\hat{\rho}_0^{\text{tot}} = Q \rho_\beta Q^\dagger \quad (3.13)$$

where

$$\rho_\beta \equiv \frac{1}{Z_\beta} e^{-\beta H_0} \quad (3.14)$$

is the initial canonical density matrix. $\beta = 1/k_B T$ is the inverse temperature and $Z_\beta = \text{Tr}(e^{-\beta H_0})$ is the corresponding partition function of the entire system. Here Q is an operator acting only on the open system. Although this operator may be chosen

in various ways to reflect specific initial conditions, here we restrict ourselves to the system initially being in full thermal equilibrium, i.e. we choose $Q = 1$.

$$\hat{\rho}_0^{\text{tot}} = \hat{\rho}_\beta \quad (3.15)$$

Having specified the initial conditions, the goal is now to derive an equation of motion that will describe the *exact* evolution of the reduced density matrix $\rho_t(q, q')$ as given by Eq. (3.12). To do this we will utilise the influence functional to eliminate the environmental degrees of freedom in Eq. (3.12).

3.2 Representation with Influence Functional

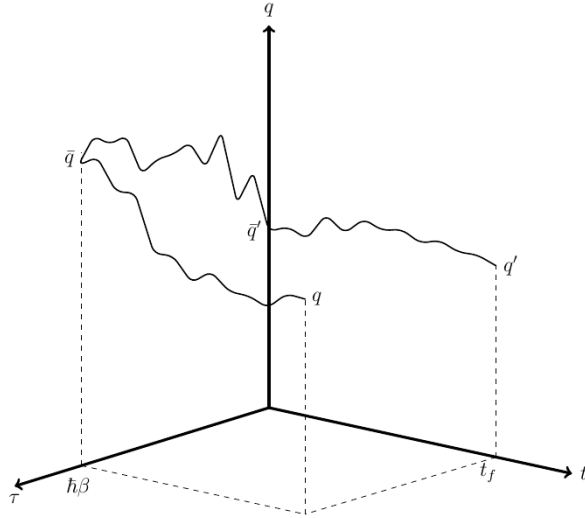


Figure 3.1: Example trajectory for the path integral in Eq. (3.16). Expressing the canonical density matrix as a path integral introduces a second time dimension, with dummy end-points \bar{q} , \bar{q}' .

In order to evaluate Eq. (3.12) we will employ the influence functional formalism outlined in section 2.1.8. The crucial generalisation is to represent both the propagators *and* the initial density matrix as path integrals. The influence functional will now

also incorporate the environmental trajectories of the imaginary time path integral and the reduced density matrix is expressible as:

$$\begin{aligned} \rho_{t_f}(q; q') &= \frac{1}{Z} \int d\bar{q} d\bar{q}' \mathcal{D}q(t) \mathcal{D}\bar{q}(\tau) \mathcal{D}q'(t) \mathcal{F}[q(t), q'(t), \bar{q}(\tau)] \\ &\times \exp\left(\frac{i}{\hbar} S_Q[q(t)] - \frac{i}{\hbar} S_Q[q'(t)] - \frac{1}{\hbar} S_Q^E[\bar{q}(\tau)]\right) \end{aligned} \quad (3.16)$$

where S_Q is the action associated with H_Q . Fig. 3.1 shows an example trajectory for this path integral in the two time dimensions. In the limiting case of no interactions we would like the influence functional to be unity. The equation above therefore uses the partition function $Z = Z_\beta/Z_B$ to normalise the influence functional in this limit. Z_B is the partition function of the (isolated) environment:

$$Z_B = \prod_{\lambda} \frac{1}{2 \sinh\left(\frac{1}{2} \omega_{\lambda} \hbar \beta\right)} \quad (3.17)$$

The new normalising constant Z in the equilibrium density operator is not generally known, and this issue will be discussed in Section 3.4.

3.2.1 Evaluating The Environmental Path Integral

The influence functional consists of the environmental and interactive contributions from H_{tot} . These have the form of a set of displaced harmonic oscillators in the environment variables, meaning the path integral in the influence functional is a trivial generalisation of Eqs. (2.81) and (2.91). It is fully factorised over the normal modes λ , and for each mode it is composed of a product of three terms, which are the contributions from the forward, backward and Euclidean propagators. Abbreviating the functions $g_{\lambda}(q(t)) = g_{\lambda}(t)$, $g_{\lambda}(q'(t)) = g'_{\lambda}(t)$ and $g_{\lambda}(\bar{q}(t)) = \bar{g}_{\lambda}(t)$, the full

influence functional is given by:

$$\mathcal{F}[q(t), q'(t), \bar{q}(\tau)] = \frac{1}{Z_B} \prod_{\lambda} \left(\frac{\omega_{\lambda}}{2\pi\hbar} \right)^{\frac{3}{2}} (\sin(\omega_{\lambda} t_f) \sinh(\omega_{\lambda} \hbar \beta))^{-\frac{1}{2}} \tilde{F}_{\lambda} \quad (3.18)$$

$$\tilde{F}_{\lambda} = \int dx_{\lambda} d\bar{x}_{\lambda} d\bar{x}'_{\lambda} F_{\lambda}[q_{\lambda}(t), x_{\lambda}, \bar{x}_{\lambda}] F_{\lambda}^E[\bar{q}_{\lambda}(\tau), \bar{x}_{\lambda}, \bar{x}'_{\lambda}] F_{\lambda}^*[q'_{\lambda}(t), x_{\lambda}, \bar{x}'_{\lambda}] \quad (3.19)$$

$$\begin{aligned} F_{\lambda}[q_{\lambda}(t), x_{\lambda}, \bar{x}_{\lambda}] = \exp \left\{ \frac{i\omega_{\lambda}}{\hbar \sin(\omega_{\lambda} t_f)} \left[\frac{1}{2} (x_{\lambda}^2 + \bar{x}_{\lambda}^2) \cos(\omega_{\lambda} t_f) - x_{\lambda} \bar{x}_{\lambda} \right. \right. \\ \left. \left. + \frac{x_{\lambda}}{\omega_{\lambda}} \int_0^{t_f} dt g_{\lambda}(t) \sin(\omega_{\lambda} t) + \frac{\bar{x}_{\lambda}}{\omega_{\lambda}} \int_0^{t_f} dt g_{\lambda}(t) \sin(\omega_{\lambda} (t_f - t)) \right. \right. \\ \left. \left. - \frac{1}{\omega_{\lambda}^2} \int_0^{t_f} \int_0^t dt dt' g_{\lambda}(t) g_{\lambda}(t') \sin(\omega_{\lambda} (t_f - t)) \sin(\omega_{\lambda} t') \right] \right\} \quad (3.20) \end{aligned}$$

$$\begin{aligned} F_{\lambda}^*[q'_{\lambda}(t), x_{\lambda}, \bar{x}'_{\lambda}] = \exp \left\{ -\frac{i\omega_{\lambda}}{\hbar \sin(\omega_{\lambda} t_f)} \left[\frac{1}{2} (x_{\lambda}^2 + \bar{x}'_{\lambda}{}^2) \cos(\omega_{\lambda} t_f) - x_{\lambda} \bar{x}'_{\lambda} \right. \right. \\ \left. \left. + \frac{x_{\lambda}}{\omega_{\lambda}} \int_0^{t_f} dt g'_{\lambda}(t) \sin(\omega_{\lambda} t) + \frac{\bar{x}'_{\lambda}}{\omega_{\lambda}} \int_0^{t_f} dt g'_{\lambda}(t) \sin(\omega_{\lambda} (t_f - t)) \right. \right. \\ \left. \left. - \frac{1}{\omega_{\lambda}^2} \int_0^{t_f} \int_0^t dt dt' g'_{\lambda}(t) g'_{\lambda}(t') \sin(\omega_{\lambda} (t_f - t)) \sin(\omega_{\lambda} t') \right] \right\} \quad (3.21) \end{aligned}$$

$$\begin{aligned} F_{\lambda}^E[\bar{q}_{\lambda}(\tau), \bar{x}_{\lambda}, \bar{x}'_{\lambda}] = \exp \left\{ -\frac{\omega_{\lambda}}{\hbar \sinh(\omega_{\lambda} \hbar \beta)} \left[\frac{1}{2} (\bar{x}_{\lambda}^2 + \bar{x}'_{\lambda}{}^2) \cosh(\omega_{\lambda} \hbar \beta) - \bar{x}_{\lambda} \bar{x}'_{\lambda} \right. \right. \\ \left. \left. - \frac{\bar{x}_{\lambda}}{\omega_{\lambda}} \int_0^{\hbar \beta} d\tau \bar{g}_{\lambda}(\tau) \sinh(\omega_{\lambda} \tau) - \frac{\bar{x}'_{\lambda}}{\omega_{\lambda}} \int_0^{\hbar \beta} d\tau \bar{g}_{\lambda}(\tau) \sinh(\omega_{\lambda} (\hbar \beta - \tau)) \right] \right\} \end{aligned}$$

$$-\frac{1}{\omega_\lambda^2} \int_0^{\hbar\beta} \int_0^\tau d\tau d\tau' \bar{g}_\lambda(\tau) \bar{g}_\lambda(\tau') \sinh(\omega_\lambda(\hbar\beta - \tau)) \sin(\omega_\lambda\tau') \Big] \Big\} \quad (3.22)$$

3.2.2 Integrating Over Environment Terminals

In order to use the influence functional, it is still necessary to perform the integration¹ over the terminals x_λ , \bar{x}_λ and \bar{x}'_λ . Despite its formidable appearance, Eq. (3.19) is simply an exponential quadratic polynomial in the integration variables. As a Gaussian integral, this is most efficiently evaluated by recasting Eq. (3.19) as a vector integral:

$$\tilde{F}_\lambda = \int d\mathbf{x} \exp \left[-\frac{1}{\hbar} (\mathbf{x}^T \boldsymbol{\Sigma} \mathbf{x} + \mathbf{b}^T \mathbf{x} + c) \right] = (\hbar\pi)^{3/2} |\boldsymbol{\Sigma}|^{-1/2} \exp \left(-\frac{1}{\hbar} \left(-\frac{1}{4} \mathbf{b}^T \boldsymbol{\Sigma} \mathbf{b} + c \right) \right) \quad (3.23)$$

$$\mathbf{x} = \begin{pmatrix} x \\ \bar{x} \\ \bar{x}' \end{pmatrix}, \quad \boldsymbol{\Sigma} = \begin{pmatrix} 0 & B & -B \\ B & \bar{A} + A & -\bar{B} \\ -B & -\bar{B} & \bar{A} - A \end{pmatrix} \quad (3.24)$$

$$\mathbf{b} = \begin{pmatrix} C[g'_\lambda - g_\lambda] \\ \bar{C}[\bar{g}_\lambda] + \bar{D}[\bar{g}_\lambda] \\ D[g'_\lambda] - \bar{D}[\bar{g}_\lambda] \end{pmatrix} \quad (3.25)$$

$$c = E[g_\lambda] - E[g'_\lambda] - \bar{E}[\bar{g}_\lambda] \quad (3.26)$$

Here the parameters in this integral are:

¹Undoubtedly the most painful calculation contained in this document.

$$A = \frac{i\omega_\lambda}{2} \cot(\omega_\lambda t_f), \quad \bar{A} = \frac{\omega_\lambda}{2} \coth(\omega_\lambda \hbar\beta) \quad (3.27)$$

$$B = i \frac{\omega_\lambda}{2 \sin(\omega_\lambda t_f)}, \quad \bar{B} = \frac{\omega_\lambda}{2 \sinh(\omega_\lambda \hbar\beta)} \quad (3.28)$$

$$C[g_\lambda] = \frac{i}{\sin(\omega_\lambda t_f)} \int_0^{t_f} dt \, g_\lambda(t) \sin(\omega_\lambda t) \quad (3.29)$$

$$D[g_\lambda] = \frac{i}{\sin(\omega_\lambda t_f)} \int_0^{t_f} dt \, g_\lambda(t) \sin(\omega_\lambda(t_f - t)) \quad (3.30)$$

$$\bar{C}[\bar{g}_\lambda] = -\frac{1}{\sinh(\omega_\lambda \hbar\beta)} \int_0^{\hbar\beta} d\tau \, \bar{g}_\lambda(\tau) \sinh(\omega_\lambda \tau) \quad (3.31)$$

$$\bar{D}[\bar{g}_\lambda] = -\frac{1}{\sinh(\omega_\lambda \hbar\beta)} \int_0^{\hbar\beta} d\tau \, \bar{g}_\lambda(\tau) \sinh(\omega_\lambda(\hbar\beta - \tau)) \quad (3.32)$$

$$E[g_\lambda] = \frac{i}{\omega_\lambda \sin(\omega_\lambda t_f)} \int_0^{t_f} \int_0^t dt dt' \, g_\lambda(t) g_\lambda(t') \sin(\omega_\lambda(t_f - t)) \sin(\omega_\lambda t') \quad (3.33)$$

$$\bar{E}[\bar{g}_\lambda] = \frac{1}{\omega_\lambda \sinh(\omega_\lambda \hbar\beta)} \int_0^{\hbar\beta} \int_0^\tau d\tau d\tau' \, \bar{g}_\lambda(\tau) \bar{g}_\lambda(\tau') \sinh(\omega_\lambda(\hbar\beta - \tau)) \sin(\omega_\lambda \tau') \quad (3.34)$$

Having recast the influence functional in this form, it is relatively easy to determine the overall prefactor. The determinant in Eq. (3.23) is easily evaluated as:

$$|\Sigma| = 2B^2 (\bar{B} - \bar{A}) = -\frac{\omega_\lambda^3 \sinh^2(\frac{1}{2}\omega_\lambda \hbar\beta)}{2 \sinh(\omega_\lambda \hbar\beta) \sin^2(\omega_\lambda t_f)}. \quad (3.35)$$

The overall prefactor for one mode of the influence functional is therefore

$$\left(\frac{\omega_\lambda}{2\pi\hbar}\right)^{\frac{3}{2}} (\sin(\omega_\lambda t_f) \sinh(\omega_\lambda \hbar\beta))^{-\frac{1}{2}} (\hbar\pi)^{3/2} |\Sigma|^{-1/2} = \frac{1}{2 \sinh(\frac{1}{2}\omega_\lambda \hbar\beta)}. \quad (3.36)$$

When taking the product of this prefactor over each mode, we recover the isolated partition function Z_B of Eq. (3.17). This cancels with the $\frac{1}{Z_B}$ in Eq. (3.18), giving an

overall unit prefactor as stipulated previously. After this cancellation, the influence functional may be represented in a purely exponential form:

$$\mathcal{F}[q, q', \bar{q}] = \exp\left(-\frac{1}{\hbar}\Phi[q, q', \bar{q}]\right) \equiv \exp\left(-\frac{1}{\hbar}\sum_{\lambda}\Phi_{\lambda}[q, q', \bar{q}]\right) \quad (3.37)$$

$$\Phi_{\lambda} = -\frac{1}{4}\mathbf{b}^T\mathbf{\Sigma}\mathbf{b} + c. \quad (3.38)$$

3.2.3 The Influence Phase

While Eq. (3.37) represents an in-principle complete evaluation of the influence functional, the form of the influence phase, $\Phi = \sum_{\lambda}\Phi_{\lambda}$ requires significant reorganisation. This is simply the exploitation of both trigonometric identities to obtain a more compact representation, but is frankly beyond tedious. After grouping terms, we obtain [57, 64]:

$$\begin{aligned} \Phi_{\lambda}[q, q', \bar{q}] = & -\int_0^{\hbar\beta} d\tau \int_0^{\tau} d\tau' K_{\lambda}(i\tau' - i\tau) \bar{g}_{\lambda}(\tau) \bar{g}_{\lambda}(\tau') \\ & -i \int_0^{\hbar\beta} d\tau \int_0^{t_f} dt K_{\lambda}(t - i\tau) \bar{g}_{\lambda}(\tau) (g_{\lambda}(t) - g'_{\lambda}(t)) \\ & + \int_0^{t_f} dt \int_0^t dt' (g_{\lambda}(t) - g'_{\lambda}(t)) [K_{\lambda}(t - t') g_{\lambda}(t') - K_{\lambda}^*(t - t') g'_{\lambda}(t')]. \end{aligned} \quad (3.39)$$

The term multiplying the various g_{λ} within the integrals is the kernel:

$$K_{\lambda}(\theta) = \frac{\cosh\left(\omega_{\lambda}\left(\frac{\hbar\beta}{2} - i\theta\right)\right)}{2\omega_{\lambda}\sinh\left(\frac{1}{2}\beta\hbar\omega_{\lambda}\right)}. \quad (3.40)$$

Note that the kernel appears in three forms, depending on purely imaginary times, $K_{\lambda}(i\tau' - i\tau)$, real times, $K_{\lambda}(t - t')$, and complex times, $K_{\lambda}(t - i\tau)$. It will be useful later in the derivation to split the kernel into its real K_{λ}^R and imaginary K_{λ}^I parts.

For real times this produces

$$K_{\lambda}^R(t) = \frac{1}{2\omega_{\lambda}} \coth\left(\frac{1}{2}\hbar\beta\omega_{\lambda}\right) \cos(\omega_{\lambda}t) \quad (3.41)$$

$$K_{\lambda}^I(t) = -\frac{1}{2\omega_{\lambda}} \sin(\omega_{\lambda}t) \quad (3.42)$$

and for complex times

$$K_{\lambda}^R(t - i\tau) = \frac{1}{2\omega_{\lambda}} \left[\coth\left(\frac{1}{2}\omega_{\lambda}\hbar\beta\right) \cosh(\omega_{\lambda}\tau) - \sinh(\omega_{\lambda}\tau) \right] \cos(\omega_{\lambda}t) \quad (3.43)$$

$$K_{\lambda}^I(t - i\tau) = -\frac{1}{2\omega_{\lambda}} \left[\cosh(\omega_{\lambda}\tau) + \sinh(\omega_{\lambda}\tau) \coth\left(\frac{1}{2}\omega_{\lambda}\hbar\beta\right) \right] \sin(\omega_{\lambda}t). \quad (3.44)$$

For purely imaginary times the kernel is real,

$$K_{\lambda}(i\tau) = K_{\lambda}^e(\tau) + K_{\lambda}^o(\tau) \quad (3.45)$$

and consists of even and odd components:

$$K_{\lambda}^o(\tau) = \frac{1}{2\omega_{\lambda}} \sinh(\omega_{\lambda}\tau) \quad (3.46)$$

$$K_{\lambda}^e(\tau) = \frac{1}{2\omega_{\lambda}} \cosh(\omega_{\lambda}\tau) \coth\left(\frac{1}{2}\omega_{\lambda}\hbar\beta\right). \quad (3.47)$$

If for real times we also define new sum and difference interaction functions [70],

$$\epsilon_{\lambda}(t) = g_{\lambda}(t) - g'_{\lambda}(t), \quad y_{\lambda}(t) = \frac{1}{2}(g_{\lambda}(t) + g'_{\lambda}(t)) \quad (3.48)$$

and substitute these expressions into Eq. (3.39), the single mode influence phase can now be expressed as:

$$\begin{aligned}
\Phi_\lambda [q, q', \bar{q}] = & - \int_0^{\hbar\beta} d\tau \int_0^{\hbar\beta} d\tau' \frac{1}{2} [K_\lambda^e(\tau' - \tau) - K_\lambda^o(|\tau' - \tau|)] \bar{g}_\lambda(\tau) \bar{g}_\lambda(\tau') \\
& - i \int_0^{\hbar\beta} d\tau \int_0^{t_f} dt [K_\lambda^R(t - i\tau) + K_\lambda^I(t - i\tau)] \bar{g}_\lambda(\tau) \epsilon_\lambda(t) \\
& + \frac{1}{2} \int_0^{t_f} dt \int_0^{t_f} dt' K_\lambda^R(t - t') \epsilon_\lambda(t) \epsilon_\lambda(t') \\
& + 2i \int_0^{t_f} dt \int_0^{t_f} dt' [\theta(t - t') K_\lambda^I(t - t')] \epsilon_\lambda(t) y_\lambda(t'). \tag{3.49}
\end{aligned}$$

The final two terms in this expression are a generalisation of the well known Feynman-Vernon influence functional [57], with the remaining terms arising from the incorporation of a non-partitioned initial density matrix. Note that, compared to Eq. (3.39), the above expression was modified to ensure identical limits in the double integrals over the times t, t' and τ, τ' .

3.2.4 Returning To The Real Space Representation

The influence phase still contains the normal mode interaction term g_λ . Using Eq. (3.11), we can re-express the phase in terms of the original interaction given in the site representation. The normal mode transformation did not change the q coordinates themselves, so there is no difference between representations in the path integral measure or action S_q in Eq. (3.16). The system-bath interaction term contained in the influence functional *will* have a different form however, and hence the influence phase has a non-trivial alternative representation in terms of functions $f_i(t) \equiv f_i(q(t))$ rather than $g_\lambda(q(t))$. In this representation the sum and difference functions

$$s_i(t) = f_i(t) - f'_i(t) \quad \text{and} \quad r_i(t) = \frac{1}{2}(f_i(t) + f'_i(t)) \quad (3.50)$$

can conveniently be introduced, using $f'_i(t) \equiv f_i(q'(t))$. Substituting Eq. (3.11) into these, we can relate the sum and difference functions (3.48) between the normal mode and site representations:

$$s_i(t) = \frac{1}{\sqrt{m_i}} \sum_{\lambda} e_{\lambda i} \epsilon_{\lambda}(t) \quad \text{and} \quad r_i(t) = \frac{1}{\sqrt{m_i}} \sum_{\lambda} e_{\lambda i} y_{\lambda}(t). \quad (3.51)$$

The influence phase in the site representation is most easily expressed by defining new kernels from those derived using normal modes

$$L_{ij}^{R,I}(t) = \frac{1}{\sqrt{m_i m_j}} \sum_{\lambda} e_{\lambda i} e_{\lambda j} K_{\lambda}^{R,I}(t) \quad (3.52)$$

$$L_{ij}(t - i\tau) = \frac{1}{\sqrt{m_i m_j}} \sum_{\lambda} e_{\lambda i} e_{\lambda j} K_{\lambda}(t - i\tau) \quad (3.53)$$

$$L_{ij}^e(\tau) = \frac{1}{\sqrt{m_i m_j}} \sum_{\lambda} e_{\lambda i} e_{\lambda j} K_{\lambda}^e(\tau) \quad (3.54)$$

$$L_{ij}^o(\tau) = \frac{1}{\sqrt{m_i m_j}} \sum_{\lambda} e_{\lambda i} e_{\lambda j} K_{\lambda}^o(\tau) \quad (3.55)$$

so that the influence phase can be re-expressed in terms of the site interactions:

$$\Phi[q, q', \bar{q}] = \sum_{ij} \Phi_{ij}[q, q', \bar{q}] \quad (3.56)$$

$$\Phi_{ij}[q, q', \bar{q}] = - \int_0^{\hbar\beta} d\tau \int_0^{\hbar\beta} d\tau' \frac{1}{2} \bar{f}_i(\tau) [L_{ij}^e(\tau' - \tau) - L_{ij}^o(|\tau' - \tau|)] \bar{f}_j(\tau')$$

$$\begin{aligned}
& -i \int_0^{\hbar\beta} d\tau \int_0^{t_f} dt s_i(t) L_{ij}(t - i\tau) \bar{f}_j(\tau) \\
& + \frac{1}{2} \int_0^{t_f} dt \int_0^{t_f} dt' s_i(t) L_{ij}^R(t - t') s_j(t') \\
& + 2i \int_0^{t_f} dt \int_0^{t_f} dt' s_i(t) [\theta(t - t') L_{ij}^I(t - t')] r_j(t')
\end{aligned} \tag{3.57}$$

where an obvious short-hand notation $f(\bar{q}(\tau)) \equiv \bar{f}_i(\tau)$ has also been introduced.

The influence phase expressed here contains additional complexity compared to one derived using a standard CL model (which does not require a normal mode transformation) [64]. After allowing the environment to contain internal couplings, we find that the effect of this generalisation on the form of the influence phase is not trivial: instead of a single sum over the bath lattice in the CL model, we have double sums in Eq. (3.56), and this will have a profound effect on the dimensionality of the stochastic field to be introduced below.

In principle, having found the influence phase, Eq. (3.16) can be used to describe the exact dynamics of the open system at all times. Path integrals are however awkward to evaluate outside of certain special cases. Instead, we will use Eq. (3.16) to derive an operator expression, and hence a Liouville-von Neumann type equation for the reduced density matrix instead². Unfortunately the influence phase in its current form presents two obstacles. First, there are double integrals in two time variables (t and τ), meaning a non-local differential equation. More problematically, there is no way to disentangle the influence functional coordinates such that the open system density matrix can be described as the product of forward, backward

²This is why the use of path integrals here is a little perverse. They are only used to reformulate the analytically tractable part of the Hamiltonian before reverting back to standard methods. Path integrals are useful, but mostly as an intermediate result- the last thing you'd want to do is actually perform a calculation with them.

and Euclidean propagators. Consequently, there is no simple method to construct a differential equation directly out of Eq. (3.16). Instead we will generalise previous work [28, 70, 71, 85], and use a transformation to convert this non-local (in the sense of the action being a double integral over time) system into a local one exactly, at the cost of introducing stochastic variables.

3.3 The Two-Time Hubbard-Stratonovich Transformation

At this point we are in a position to use the results of section 2.2.5. For our purposes, we require two time dimensions, t and τ , running up to t_f and $\hbar\beta$ respectively, with associated stochastic fields $z(t)$ and $\bar{z}(\tau)$. Here we will indicate the average over the noise realisations $\{z(t), \bar{z}(\tau)\}$ with $\langle \dots \rangle_r$. In this case, Eq. (2.167) reduces to:

$$\begin{aligned}
& \left\langle \exp \left[i \sum_{i\alpha} \left(\int_0^{t_f} dt \, z_i^\alpha(t) k_i^\alpha(t) + \int_0^{\hbar\beta} d\tau \, \bar{z}_i^\alpha(\tau) \bar{k}_i^\alpha(\tau) \right) \right] \right\rangle_r \\
&= \exp \left[-\frac{1}{2} \sum_{ij\alpha\beta} \left(\int_0^{t_f} dt \int_0^{t_f} dt' \, k_i^\alpha(t) A_{ij}^{\alpha\beta}(t, t') k_j^\beta(t') \right. \right. \\
&+ \left. \int_0^{\hbar\beta} d\tau \int_0^{\hbar\beta} d\tau' \, \bar{k}_i^\alpha(\tau) A_{ij}^{\alpha\beta}(\tau, \tau') \bar{k}_j^\beta(\tau') + 2 \int_0^{t_f} dt \int_0^{\hbar\beta} d\tau \, k_i^\alpha(t) A_{ij}^{\alpha\beta}(t, \tau) \bar{k}_j^\beta(\tau) \right) \Big] \\
& \tag{3.58}
\end{aligned}$$

$$A_{ij}^{\alpha\beta}(t, t') = \left\langle z_i^\alpha(t) z_j^\beta(t') \right\rangle_r \tag{3.59}$$

$$A_{ij}^{\alpha\beta}(\tau, \tau') = \left\langle \bar{z}_i^\alpha(\tau) \bar{z}_j^\beta(\tau') \right\rangle_r \tag{3.60}$$

$$A_{ij}^{\alpha\beta}(t, \tau) = \left\langle z_i^\alpha(t) \bar{z}_j^\beta(\tau) \right\rangle_r \quad (3.61)$$

The connection between the influence phase in Eq. (3.57) and the exponent in Eq. (3.58) should become transparent with the choice of t_f or $\hbar\beta$ as the upper limits of each time integration. The choice for the second time dimension to run up to $\hbar\beta$ has been made to highlight the closeness between the influence phase in Eq. (3.57) and the two-time HS transformation presented here.

Now we would like to apply the HS transformation to the influence functional expression given by Eqs. (3.37), (3.56) and (3.57). It is clear from the structure of the exponent in the influence functional in Eq. (3.57), that auxiliary stochastic fields should be introduced separately for each lattice site index i . Moreover, there should be two pairs of the stochastic processes for the set associated with the real time t ,

$$z_i(t) \Rightarrow \begin{pmatrix} \eta_i(t) \\ \eta_i^*(t) \\ \nu_i(t) \\ \nu_i^*(t) \end{pmatrix} \quad (3.62)$$

and one such set for the imaginary time $i\tau$:

$$\bar{z}_i(\tau) \Rightarrow \begin{pmatrix} \bar{\mu}_i(\tau) \\ \bar{\mu}_i^*(\tau) \end{pmatrix} \quad (3.63)$$

where we have redefined the complex vector $z \equiv \{z_i\}$ to include two noises and their conjugates. Next, we make the following correspondence between the functions $k_i(t)$ in the HS transformation (3.58) and the functions $s_i(t)$, $r_i(t)$ and $\bar{f}_i(\tau)$ appearing in

the phase, Eq. (3.57):

$$k_i(t) \Rightarrow \begin{pmatrix} \frac{s_i(t)}{\hbar} \\ 0 \\ r_i(t) \\ 0 \end{pmatrix} \quad (3.64)$$

and

$$\bar{k}_i(\tau) \Rightarrow \begin{pmatrix} \frac{i\bar{f}_i(\tau)}{\hbar} \\ 0 \end{pmatrix}. \quad (3.65)$$

The three pairs of stochastic processes we have introduced must ensure that the influence functional given by Eqs. (3.37), (3.56) and (3.57) coincide exactly with the right hand side of the HS transformation (3.58). Therefore, comparing the exponents in the right hand side of Eq. (3.58) and Eq. (3.57), explicit formulas can be established for the correlation functions $A_{ij}^{\alpha\beta}$ between the noises. These are:

$$\langle \eta_i(t) \eta_j(t') \rangle_r = \hbar L_{ij}^R(t - t') \quad (3.66)$$

$$\langle \eta_i(t) \nu_j(t') \rangle_r = 2i\Theta(t - t') L_{ij}^I(t - t') \quad (3.67)$$

$$\langle \eta_i(t) \bar{\mu}_j(\tau) \rangle_r = -\hbar [L_{ij}^R(t - i\tau) + iL_{ij}^I(t - i\tau)] \quad (3.68)$$

$$\langle \bar{\mu}_i(\tau) \bar{\mu}_j(\tau') \rangle_r = \hbar [L_{ij}^e(\tau - \tau') - L_{ij}^o(|\tau - \tau'|)] \quad (3.69)$$

$$\langle \nu_i(t) \nu_j(t') \rangle_r = \langle \nu_i(t) \bar{\mu}_j(\tau) \rangle_r = 0. \quad (3.70)$$

Note that the correlation functions (3.66) and (3.69) are to be symmetric functions with respect to the permutation $i, t \leftrightarrow j, t'$ and $i, \tau \leftrightarrow j, \tau'$, respectively, and the corresponding functions L_{ij}^R and $L_{ij}^{o,e}$ provide exactly this.

Taking the above results and applying them to Eq. (3.57), we find that the influence functional can be described as an average over multivariate complex Gaussian processes as follows:

$$\mathcal{F}[q, q', \bar{q}] = \left\langle \exp \left[\frac{i}{\hbar} \sum_i \left(\int_0^{t_f} dt [\eta_i(t) s_i(t) + \hbar \nu_i(t) r_i(t)] + i \int_0^{\hbar\beta} d\tau \bar{\mu}_i(\tau) \bar{f}_i(\tau) \right) \right] \right\rangle_r \quad (3.71)$$

where the averaging is made over three pairs of complex noises (or, equivalently, over six real noises) per lattice site of the environment.

Importantly, the two-time HS transformation is a purely formal one, and we are free to stipulate that the noises are pure c -numbers. This enables us to avoid the complication of operator-valued noises, which has been previously shown to have no material effect on the final result [70, 71].

Finally it is worth mentioning that the influence phase given above does *not* uniquely define the Gaussian processes that the influence functional is averaged over after performing the mapping. The influence phase viewed as the right hand side of the HS transformation does not involve every possible correlation defined under the Gaussian distribution. In particular, the conditions we impose on some correlation functions to map the physics to the auxiliary noises do not constrain the correlations between the complex conjugate noises, e.g. $\langle \eta_i^*(t) \eta_j^*(t') \rangle$. Therefore any distribution that satisfies Eqs. (3.66)-(3.70) may be used in this transformation.

3.4 The Extended Stochastic Liouville-von Neumann Equation

Now the influence functional $\mathcal{F}[q, q', \bar{q}]$ has (finally) been evaluated, we are able to give a new explicit representation for the reduced density matrix in Eq. (3.16). First,

having introduced stochastic variables into the equation for the density matrix, we must define a new object $\tilde{\rho}_t(q; q')$ to act as an effective, single-trajectory density matrix defined for a particular realisation of the stochastic processes $z(t)$ and $\bar{z}(\tau)$ along its path. Inserting Eq. (3.71) into Eq.(3.16) we obtain,

$$\tilde{\rho}_{t_f}(q; q') = \frac{1}{Z} \int d\bar{q} d\bar{q}' \mathcal{D}q(t) \mathcal{D}\bar{q}(\tau) \mathcal{D}q'(t) \exp \left[\frac{i}{\hbar} \tilde{S}^+[q(t)] - \frac{i}{\hbar} \tilde{S}^-[q'(t)] - \frac{1}{\hbar} \tilde{S}^E[\bar{q}(\tau)] \right] \quad (3.72)$$

where three effective actions have been introduced:

$$\tilde{S}^+[q(t)] = \int_0^{t_f} dt \left(L_q[q(t)] + \sum_i \left[\eta_i(t) + \frac{\hbar}{2} \nu_i(t) \right] f_i(t) \right) = \int_0^{t_f} dt L^+[q(t)] \quad (3.73)$$

$$\tilde{S}^-[q'(t)] = \int_0^{t_f} dt \left(L_q[q'(t)] + \sum_i \left[\eta_i(t) - \frac{\hbar}{2} \nu_i(t) \right] f_i(t) \right) = \int_0^{t_f} dt L^-[q'(t)] \quad (3.74)$$

$$\tilde{S}^E[\bar{q}(\tau)] = \int_0^{\hbar\beta} d\tau \left(H_Q[\bar{q}(\tau)] + \bar{\mu}_i(\tau) \bar{f}_i(\tau) \right). \quad (3.75)$$

The physical density matrix is recovered by averaging over $\tilde{\rho}_t(q; q')$:

$$\rho_{t_f}(q; q') = \langle \tilde{\rho}_{t_f}(q; q') \rangle_{\{z(t), \bar{z}(\tau)\}}. \quad (3.76)$$

In the definitions of the effective actions we have reinserted the original forces $f_i(t)$, $f_i(t')$ and $\bar{f}_i(\tau)$ via Eq. (3.50). It can be seen that the actions \tilde{S}^+ and \tilde{S}^- correspond to two different effective Lagrangians,

$$\hat{L}^\pm(t) = \hat{L}_Q(t) + \sum_i \left[\eta_i(t) \pm \frac{\hbar}{2} \nu_i(t) \right] \hat{f}_i(t) \quad (3.77)$$

which in turn are associated with two different effective Hamiltonians:

$$\hat{H}^{\pm}(t) = \hat{H}_Q(t) - \sum_i \left[\eta_i(t) \pm \frac{\hbar}{2} \nu_i(t) \right] \hat{f}_i(t). \quad (3.78)$$

As was mentioned in Section 3.3, the noises are not promoted to operators but remain as c -numbers.

3.4.1 Effective Propagators

All three path integral coordinates have now been decoupled from each other, and as coordinate functionals may be commuted. The density matrix in Eq. (3.72) can therefore be expressed as:

$$\tilde{\rho}_{t_f}(q; q') = \int d\bar{q} d\bar{q}' U^+(q, t_f; \bar{q}, 0) \tilde{\rho}_0(\bar{q}; \bar{q}') U^-(\bar{q}', 0; q', t_f) \equiv \langle q | \tilde{\rho}(t_f) | q' \rangle \quad (3.79)$$

where

$$U^+(q, t_f; \bar{q}, 0) = \int_{q(0)=\bar{q}}^{q(t_f)=q} \mathcal{D}q(t) \exp \left[\frac{i}{\hbar} \tilde{S}^+[q(t)] \right] \equiv \langle q | \hat{U}^+(t_f) | \bar{q} \rangle \quad (3.80)$$

$$U^-(\bar{q}', 0; q', t_f) = \int_{q'(t_f)=q'}^{\bar{q}'(0)=\bar{q}'} \mathcal{D}q'(t) \exp \left[-\frac{i}{\hbar} \tilde{S}^-[q'(t)] \right] \equiv \langle \bar{q}' | \hat{U}^-(t_f) | q' \rangle \quad (3.81)$$

$$\tilde{\rho}_0(\bar{q}; \bar{q}') = \frac{1}{Z} \int_{\bar{q}(0)=\bar{q}'}^{\bar{q}(\hbar\beta)=\bar{q}} \mathcal{D}\bar{q}(\tau) \exp \left[-\frac{1}{\hbar} \tilde{S}^E[\bar{q}(\tau)] \right] \equiv \langle \bar{q} | \tilde{\rho}_0 | \bar{q}' \rangle. \quad (3.82)$$

Notice that the forwards propagator is *not* the Hermitian conjugate of the backwards propagator because of the obvious difference in their respective Hamiltonians. The consequence of this is that the equation of motion is no longer of the Liouville form,

i.e. the time derivative of the density matrix is not solely given by the commutator with some kind of Hamiltonian.

Within Eqs. (3.80) and (3.81) we have also introduced the operators

$$\hat{U}^+(t_f) = \hat{T} \exp \left(-\frac{i}{\hbar} \int_0^{t_f} \hat{H}^+(t) dt \right) \quad (3.83)$$

$$\hat{U}^-(t_f) = \tilde{T} \exp \left(\frac{i}{\hbar} \int_0^{t_f} \hat{H}^-(t) dt \right) \quad (3.84)$$

which correspond to the forward and backward propagation performed with the different Hamiltonians \hat{H}^+ and \hat{H}^- respectively, using the corresponding chronological \hat{T} and anti-chronological \tilde{T} time-ordering operators. It is easy to see that the coordinate representation $\langle q | \hat{U}^+(t_f) | \bar{q} \rangle$ and $\langle \bar{q}' | \hat{U}^-(t_f) | q' \rangle$ of such operators give exactly the paths integrals in these expressions. The propagator operators satisfy the usual equations of motion

$$i\hbar \partial_t \hat{U}^+(t) = \hat{H}^+(t) \hat{U}^+(t) \quad (3.85)$$

$$i\hbar \partial_t \hat{U}^-(t) = -\hat{U}^-(t) \hat{H}^-(t). \quad (3.86)$$

3.4.2 Real time Evolution

Taking Eqs. (3.79)-(3.82), the reduced single-trajectory density matrix $\tilde{\rho}(t_f)$ of the open system can be written as an operator evolution:

$$\tilde{\rho}(t) = \hat{U}^+(t) \tilde{\rho}_0 \hat{U}^-(t). \quad (3.87)$$

With these definitions it is possible to generate an equation of motion for a single-trajectory reduced density matrix by simply differentiating the above expression with

respect to time:

$$\begin{aligned}
i\hbar\partial_t\tilde{\rho}(t) &= \hat{H}^+(t)\tilde{\rho}(t) - \tilde{\rho}(t)\hat{H}^-(t) \\
&= [\hat{H}_Q(t), \tilde{\rho}(t)] - \sum_i \left(\eta_i(t) [\hat{f}_i(t), \tilde{\rho}(t)] + \frac{\hbar}{2}\nu_i(t) \{ \hat{f}_i(t), \tilde{\rho}(t) \} \right) \quad (3.88)
\end{aligned}$$

At first glance this is identical (with a generalised coupling) to the equation derived by Stockburger [85] using the partitioned approach, reflecting the fact that the structure of the real time dynamics do not depend on initial conditions. The crucial difference is that in the extended case the noises must satisfy extra correlations, tying them to the imaginary time evolution.

We also note that, as was mentioned above, the obtained equation does not have the usual Liouville form because of an extra anti-commutator term in the right hand side. This originates from the fact that the forward and backward propagations of the reduced density matrix in Eq. (3.87), are governed by different Hamiltonians. The same equation of motion for the reduced density matrix can also be obtained using the method developed by Kleinert and Shabanov in Ref. [70], however their method requires some care in choosing the correct order of the coordinates and momenta operators, which is a redundant consideration in this treatment.

3.4.3 Imaginary Time Evolution

All that remains is to determine the new single-trajectory initial density matrix $\tilde{\rho}_0$. This is the true initial ($t = 0$) single-trajectory reduced density matrix which is obtained from the canonical density matrix (3.15) by tracing out the degrees of freedom of the bath. There is already a path integral representation for this density, Eq. (3.82), but it is unwieldy and unintuitive. Once again it is best to work backwards

to obtain the corresponding effective canonical initial density matrix operator $\tilde{\rho}_0$ with the same path integral representation. We would like to work backwards to obtain the effective canonical Hamiltonian for the initial density matrix, in the same way as we did for the propagators. This is not straightforward due to the imaginary time dependence on the noises. In order to proceed we shall have to postulate what we term a quenched canonical density matrix:

$$\bar{\rho}(\hbar\beta) = \frac{1}{Z} \hat{\tau} \exp \left[-\frac{1}{\hbar} \int_0^{\hbar\beta} d\tau \bar{H}(\tau) \right] \quad (3.89)$$

This object bears many similarities to the propagator for a time-dependent Hamiltonian. It can be considered as the equilibrium density matrix produced from a temperature dependent Hamiltonian, when one quenches from infinite temperature, down to the desired temperature. The $\hat{\tau}$ operator orders inverse-temperatures, in much the same way as the time ordering operator does for propagators. We shall now demonstrate that the path integral representation of this density matrix has the form of Eq.(3.82).

The quenched density in coordinate space is:

$$\bar{\rho}(q; q') = \langle q | \bar{\rho} | q' \rangle \quad (3.90)$$

In order to generate the path integral, we must first discretise the operator using $\Delta N = \hbar\beta$ and $\bar{H}_j = \bar{H}(j\Delta)$:

$$\exp \left[-\frac{1}{\hbar} \int_0^{\hbar\beta} d\tau \bar{H}(\tau) \right] \rightarrow \exp \left[-\frac{1}{\hbar} \sum_{j=0}^N \Delta \bar{H}_j \right]. \quad (3.91)$$

We know that we will take the limit $\Delta \rightarrow 0$ to obtain the path integral, so we will apply the Zassenhaus approximation to our discretised density. This approximation states

$$\exp \left[\Delta \left(\hat{A} + \hat{B} \right) \right] = \exp \left[\Delta \hat{A} \right] \exp \left[\Delta \hat{B} \right] \exp \left[-\frac{\Delta^2}{2} \left[\hat{A}, \hat{B} \right]_- \right] \times \mathcal{O} \left(\Delta^3 \right) \quad (3.92)$$

which after application yields

$$Z\bar{\rho} \approx \left[\prod_{j=0}^{N-1} \exp \left[-\frac{\Delta^2}{2\hbar^2} \left[\bar{H}_{j+1}, \bar{H}_j \right]_- \right] \right] \hat{\tau} \prod_{j=0}^N \exp \left[-\frac{1}{\hbar} \Delta \bar{H}_j \right]. \quad (3.93)$$

If we assume the commutator between any two of \bar{H}_j is proportional to \hbar then we have

$$\left[\prod_{j=0}^{N-1} \exp \left[-\frac{\Delta^2}{2\hbar^2} \left[\bar{H}_{j+1}, \bar{H}_j \right]_- \right] \right] \propto \exp \left[-\frac{a\Delta^2}{2\hbar} \right]. \quad (3.94)$$

In the limit this term will become unity, so we shall neglect it and focus on the remaining part $\hat{\tau} \prod_{j=0}^N \exp \left[-\frac{1}{\hbar} \Delta \bar{H}_j \right]$. After τ ordering this the term becomes

$$\hat{\tau} \prod_{j=0}^N \exp \left[-\frac{1}{\hbar} \Delta \bar{H}_j \right] = \exp \left[-\frac{1}{\hbar} \Delta \bar{H}_N \right] \dots \exp \left[-\frac{1}{\hbar} \Delta \bar{H}_j \right] \dots \exp \left[-\frac{1}{\hbar} \Delta \bar{H}_0 \right]. \quad (3.95)$$

We can use the Zassenhaus approximation again to split the Hamiltonian into its kinetic and potential parts

$$Z\bar{\rho}(q; q') \approx \langle q | \exp \left[-\frac{1}{\hbar} \Delta \bar{T}_N \right] \exp \left[-\frac{1}{\hbar} \Delta \bar{V}_N \right] \dots \exp \left[-\frac{1}{\hbar} \Delta \bar{T}_0 \right] \exp \left[-\frac{1}{\hbar} \Delta \bar{V}_0 \right] | q' \rangle. \quad (3.96)$$

Now we insert a resolution of the identity in terms of the \bar{V}_j position eigenstates. This gives:

$$\bar{\rho}(q; q') = \int dq_1 \dots dq_n \prod_{j=0}^N \left\langle q_{j+1} \left| \exp \left[-\frac{1}{\hbar} \Delta \bar{T}_j \right] \exp \left[-\frac{1}{\hbar} \Delta \bar{V}_j \right] \right| q_j \right\rangle \quad (3.97)$$

using $|q_0\rangle = |q'\rangle$ and $|q_{N+1}\rangle = |q\rangle$.

We are now in a position to evaluate a single term in the product

$$\left\langle q_{j+1} \left| \exp \left[-\frac{1}{\hbar} \Delta \bar{T}_j \right] \exp \left[-\frac{1}{\hbar} \Delta \bar{V}_j \right] \right| q_j \right\rangle = \left\langle q_{j+1} \left| \exp \left[-\frac{1}{\hbar} \Delta \bar{T}_j \right] \right| q_j \right\rangle \exp \left[-\frac{1}{\hbar} \Delta \bar{V}(q_j, j\Delta) \right] \quad (3.98)$$

while the kinetic operator can be assessed by transforming to the momentum basis

$$\left\langle q_{j+1} \left| \exp \left[-\frac{1}{\hbar} \Delta \bar{T}_j \right] \right| q_j \right\rangle = \int dp \langle q_{j+1} | \exp \left[-\frac{1}{\hbar} \Delta \bar{T}_j \right] | p \rangle \langle q_j | p \rangle \quad (3.99)$$

$$= \int dp \langle q_{j+1} | p \rangle \langle q_j | p \rangle \exp \left[-\frac{\Delta p^2}{2m\hbar} \right] \quad (3.100)$$

resulting in the following expression for a single term:

$$\left\langle q_{j+1} \left| \exp \left[-\frac{1}{\hbar} \Delta \bar{T}_j \right] \exp \left[-\frac{1}{\hbar} \Delta \bar{V}_j \right] \right| q_j \right\rangle = \int dp \frac{1}{2\pi\hbar} \exp \left[-\frac{1}{\hbar} \left(\frac{\Delta p^2}{2m} - i(q_{j+1} - q_j) \right) \right]. \quad (3.101)$$

This can be integrated by completing the square

$$\left\langle q_{j+1} \left| \exp \left[-\frac{1}{\hbar} \Delta \bar{T}_j \right] \right| q_j \right\rangle = \sqrt{\frac{m}{2\pi\hbar\Delta}} \exp \left[-\frac{m}{2\Delta\hbar} (q_{j+1} - q_j)^2 \right] \quad (3.102)$$

and reinserted into Eq.(3.97) to produce the path integral representation for the

quenched density:

$$\bar{\rho}(q; q') = \lim_{\Delta \rightarrow 0} \frac{1}{Z} \int \prod_{j=0}^N \left(\sqrt{\frac{m}{2\pi\hbar\Delta}} dq_j \right) \exp \left[-\frac{\Delta}{\hbar} \left(\frac{m(q_{j+1} - q_j)^2}{2\Delta^2} + \bar{V}(q_j, j\Delta) \right) \right] \quad (3.103)$$

$$= \frac{1}{Z} \int \mathcal{D}\bar{q}(\tau) \exp \left[-\frac{1}{\hbar} \int_0^{\hbar\beta} d\tau \bar{H}(\tau) \right] \quad (3.104)$$

with $\bar{H}(\tau)$ being a simple function of imaginary time (not an operator).

The path integral representation of the initial density matrix in Eq. (3.82) may now be identified with Eq. (3.89), using an effective Hamiltonian given by Eq. (3.75):

$$\bar{H}(\tau) = \hat{H}_Q(0) + \sum_i^M \bar{\mu}_i(\tau) \hat{f}_i(0). \quad (3.105)$$

The initial reduced density operator is now characterised as a propagator through imaginary time:

$$\tilde{\rho}_0 \equiv \bar{\rho}(\tau)|_{\tau=\beta\hbar} \quad (3.106)$$

using

$$\bar{\rho}(\tau) = \frac{1}{Z} \hat{\tau} \exp \left[-\frac{1}{\hbar} \int_0^\tau d\tau' \left(\hat{H}_Q(0) + \sum_i^M \bar{\mu}_i(\tau') \hat{f}_i(0) \right) \right]. \quad (3.107)$$

This density operator $\bar{\rho}(\tau)$ is responsible for the thermalisation of the open system (when $\tau \rightarrow \beta\hbar$) and satisfies the Schrödinger-like equation of motion:

$$-\hbar \partial_\tau \bar{\rho}(\tau) = \bar{H}(\tau) \bar{\rho}(\tau) \quad (3.108)$$

$$\bar{\rho}(\tau=0) = Z^{-1} \quad (3.109)$$

The initial density $\bar{\rho}(\tau)$ must be normalised when the final value of $\tau \equiv \beta\hbar$ is

reached, i.e. $\text{Tr}_Q [\bar{\rho}(\beta\hbar)] = 1$, where the trace is taken with respect to the open system only. Therefore, the correct initial condition for $\bar{\rho}(\tau)$ can be fixed by providing this normalisation at the end of the imaginary time propagation (note that Z , as a ratio of two partition functions, is time independent).

The Hamiltonian H_Q and the interaction operators in $\bar{H}(\tau)$ have no temperature dependence, but instead the temperature sets the propagation “time” $\tau = \beta\hbar$. This hard limit relating the time to the system temperature is important, as unlike in the real time case, the effective initial density matrix may diverge as we take $\tau \rightarrow \infty$. This is a reflection of the fact that the path integral description of the canonical density matrix is itself only defined for finite temperature.

3.4.4 The ESLE

Taken together, the equations (3.88), (3.106) and (3.108) form the ESLE. They provide the complete solution for the real time evolution of the reduced density matrix of an open system in our partition-free approach. They are implemented by first propagating in imaginary time $\bar{\rho}(\tau)$ up to the final time $\tau \equiv \beta\hbar$ (the Euclidean evolution). The initial density is then normalised which fixes the value of the partition function Z . Using the obtained initial density matrix, the real time dynamics of the reduced density matrix $\tilde{\rho}(t)$ are elucidated by solving Eq. (3.88).

Figure 3.2 illustrates the evolution of trajectories through two times, as governed by the two differential equations. First the system evolves through imaginary time according to Eq. (3.108) and some realisation of the imaginary time noise trajectory $\{\bar{\mu}_i(\tau)\}$. This state then evolves through real time under Eq. (3.88) using the real time noise trajectories $\{\eta_i(t)\}$ and $\{\nu_i(t)\}$, with the requirement that upon averaging over realisations of these trajectories, they satisfy the correlation functions derived in section 3.3. The evolution along these two time dimensions is then repeated many times using various realisations of the stochastic noises, and averaging over many

trajectories yields the physical reduced density matrix $\hat{\rho}(t)$ appearing in Eq. (3.76).

Having finally derived this set of equations, it is natural to ask what can be done with it. A few results derivable from the ESLE will be sketched in the next section.

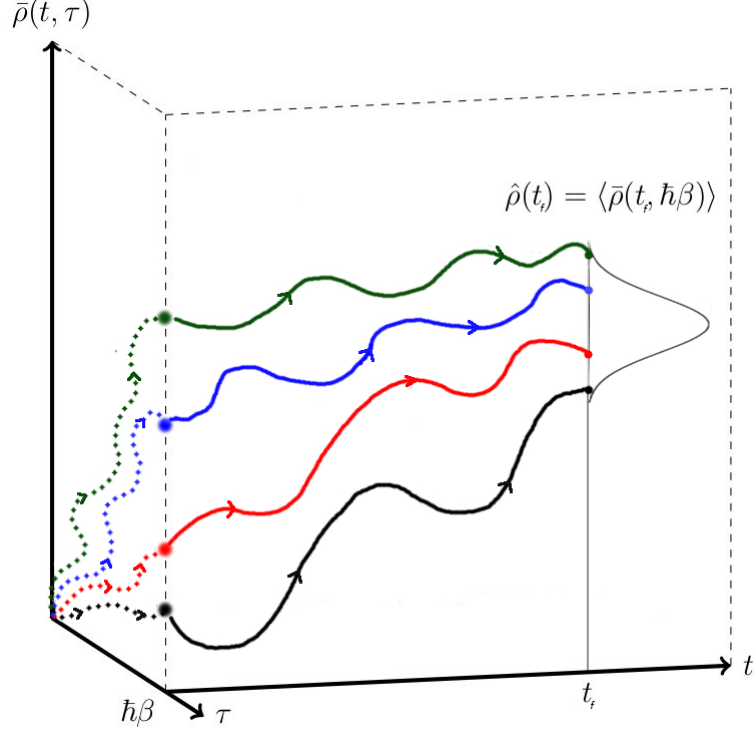


Figure 3.2: Representative trajectories for the evolution of the system. First there is an evolution in imaginary time up to $\tau = \beta\hbar$, before evolving in real time from this point up to time t_f . Different colours correspond to different simulations associated with particular manifestations of the noises. The average of the final points gives the physical density matrix at that time (indicated at time t_f).

3.5 Derivations Using the ESLE

In this section we will outline some short results that can be derived using the ESLE. These are not calculations of specific systems, but designed to illustrate generic behaviours and generalisations of the ESLE. Bearing this in mind, the first derivation will provide a simplification to reduce the dimensionality of the model.

3.5.1 A Simplified Model

The prescription considered so far requires three noises $(\eta_i(t), \nu_i(t))$ and $\bar{\mu}_i(t)$ per bath lattice site i . It is useful however when sketching essential features to employ a simpler model, using the normal modes x_λ instead of the site representation. Formally we replace in Eq. (3.1) $m_i \rightarrow 1, i \rightarrow \lambda, \Lambda_{ij} \rightarrow \omega_\lambda^2 \delta_{\lambda\lambda'}, e_{i\lambda'} \rightarrow e_{\lambda\lambda'} = \delta_{\lambda\lambda'}$ and $f_i(q) \rightarrow f_\lambda(q)$, which reduces the description to the standard Caldeira-Leggett model [11] albeit with a slightly more general coupling term, $-\sum_\lambda f_\lambda(q)\xi_\lambda$. In this representation all the correlation function matrices become diagonal, e.g.

$$L_{\lambda\lambda'}^R(t) = \delta_{\lambda\lambda'} L_{\lambda\lambda}^R(t) = \delta_{\lambda\lambda'} \frac{1}{2\omega_\lambda} \coth\left(\frac{1}{2}\omega_\lambda \hbar\beta\right) \cos(\omega_\lambda t)$$

The main simplification then comes by assuming that, up to a scaling factor, the q dependences in the coupling functions $f_\lambda(q)$ are identical, i.e. $f_\lambda(q) = c_\lambda f(q)$. In this prescription it is possible to collectively redefine the noise terms reducing them to just three distinct terms. Taking the $\eta_i \rightarrow \eta_\lambda$ noises in Eq. (3.88) as an example:

$$\sum_i \eta_i(t) [f_i(q), \tilde{\rho}(t)]_- \rightarrow \eta(t) [f(q), \tilde{\rho}(t)]_- \quad (3.110)$$

with $\eta(t) = \sum_\lambda c_\lambda \eta_\lambda(t)$ being a new Gaussian noise. The correlation function of this combined noise will be given by:

$$\begin{aligned} \langle \eta(t) \eta(t') \rangle_r &= \hbar \sum_{\lambda\lambda'} c_\lambda c_{\lambda'} L_{\lambda\lambda'}^R(t) = \hbar \sum_\lambda c_\lambda^2 L_{\lambda\lambda}^R(t) \\ &= \hbar \sum_\lambda \frac{c_\lambda^2}{2\omega_\lambda} \coth\left(\frac{1}{2}\omega_\lambda \hbar\beta\right) \cos(\omega_\lambda (t - t')) \end{aligned} \quad (3.111)$$

In the continuum limit we can replace the sum over bath modes with an integration over frequency:

$$\sum_{\lambda} \frac{c_{\lambda}^2}{2\omega_{\lambda}} \rightarrow \int_0^{\infty} \frac{d\omega}{\pi} \left[\pi \sum_{\lambda} \frac{c_{\lambda}^2}{2\omega_{\lambda}} \delta(\omega - \omega_{\lambda}) \right] = \int_0^{\infty} \frac{d\omega}{\pi} I(\omega). \quad (3.112)$$

Here $I(\omega)$ is the bath spectral density, which is formally dependent on the specific model which couples oscillators of the environment and the system.

Similarly to the $\eta(t)$ noise, two other collective noises, $\nu(t)$ and $\bar{\mu}(\tau)$, are introduced, bringing the total to three. In the reduced case, which we shall use in the rest of this section, the ESLE can be completely described by two stochastic differential equations, one for the initialisation in the imaginary time,

$$-\hbar \partial_{\tau} \bar{\rho}(\tau) = \left[\hat{H}_Q(t_0) + \bar{\mu}(\tau) \hat{f}(t_0) \right] \bar{\rho}(\tau) \quad (3.113)$$

and another for the propagation in real time:

$$i\hbar \partial_t \tilde{\rho}(t) = \left[\hat{H}_Q(t), \tilde{\rho}(t) \right] - \eta(t) \left[\hat{f}(t), \tilde{\rho}(t) \right] - \frac{\hbar}{2} \nu(t) \left\{ \hat{f}(t), \tilde{\rho}(t) \right\} \quad (3.114)$$

where the corresponding correlation functions are given by the following equations:

$$\langle \eta(t) \eta(t') \rangle_r = \hbar \int_0^{\infty} \frac{d\omega}{\pi} I(\omega) \coth\left(\frac{1}{2}\omega\hbar\beta\right) \cos(\omega(t-t')) \equiv K_{\eta\eta}(t-t') \quad (3.115)$$

$$\langle \eta(t) \nu(t') \rangle_r = -2i\Theta(t-t') \int_0^{\infty} \frac{d\omega}{\pi} I(\omega) \sin(\omega(t-t')) \equiv K_{\eta\nu}(t-t') \quad (3.116)$$

$$\langle \eta(t) \bar{\mu}(\tau) \rangle_r = -\hbar \int_0^{\infty} \frac{d\omega}{\pi} I(\omega) \frac{\cosh\left(\omega\left(\frac{\hbar\beta}{2} - \tau - it\right)\right)}{\sinh\left(\frac{1}{2}\beta\hbar\omega\right)} \equiv K_{\eta\bar{\mu}}(t-i\tau) \quad (3.117)$$

$$\langle \bar{\mu}(\tau) \bar{\mu}(\tau') \rangle_r = \hbar \int_0^\infty \frac{d\omega}{\pi} I(\omega) \frac{\cosh\left(\omega\left(\frac{\hbar\beta}{2} - \tau + \tau'\right)\right)}{\sinh\left(\frac{1}{2}\beta\hbar\omega\right)} \equiv K_{\bar{\mu}\bar{\mu}}(\tau - \tau') \quad (3.118)$$

$$\langle \nu(t) \nu(t') \rangle_r = \langle \nu(t) \bar{\mu}(\tau) \rangle_r = 0 \quad (3.119)$$

3.5.2 Classical Limit

While the ESLE is formally rather simple, it is difficult to interpret physically. The classical limit of the ESLE is useful in establishing some context. As a first pass we will take this limit heuristically, while in chapter 5 we employ the full power of KvN dynamics to make a rigorous calculation.

Focusing first on dynamics, we know that in the classical case the only path from the propagator

$$\hat{U}^\pm(t_f) = \hat{T} \exp\left(-\frac{i}{\hbar} \int_0^{t_f} \hat{L}_Q(t) - \left[\eta(t) \pm \frac{1}{2}\nu'(t)\right] f(t) dt\right) \quad (3.120)$$

that contributes is that along the path with the action $\tilde{S}_{\text{cl}}^\pm$:

$$\tilde{S}_{\text{cl}}^\pm = \int_0^{t_f} dt L_Q(q, t) - \left[\eta_{\text{cl}}(t) \pm \frac{1}{2}\nu'_{\text{cl}}(t)\right] f(q, t) \quad (3.121)$$

where we have redefined the ν noise as $\nu' = \hbar\nu$. The correlation functions for the noises will also be affected in the classical limit, hence we define new noises that obey these limiting correlation functions. The classical limit of all correlation functions can be calculated using the identity

$$\lim_{x \rightarrow 0} x \coth(ax) = \frac{1}{a}. \quad (3.122)$$

As a result, we obtain:

$$\lim_{\hbar \rightarrow 0} \langle \eta(t) \eta(t') \rangle_r = \langle \eta_{\text{cl}}(t) \eta_{\text{cl}}(t') \rangle_r = 2k_B T \int_0^\infty \frac{d\omega}{\omega\pi} I(\omega) \cos(\omega(t-t')) \quad (3.123)$$

$$\lim_{\hbar \rightarrow 0} \langle \eta(t) \bar{\mu}(\tau) \rangle_r = \langle \eta_{\text{cl}}(t) \bar{\mu}_{\text{cl}}(\tau) \rangle_r = -2k_B T \int_0^\infty \frac{d\omega}{\omega\pi} I(\omega) \cosh(\tau + it) \quad (3.124)$$

$$\lim_{\hbar \rightarrow 0} \langle \bar{\mu}(\tau) \bar{\mu}(\tau') \rangle_r = \langle \bar{\mu}_{\text{cl}}(\tau) \bar{\mu}_{\text{cl}}(\tau') \rangle_r = 2k_B T \int_0^\infty \frac{d\omega}{\omega\pi} I(\omega) \cosh(\tau - \tau') \quad (3.125)$$

$$\lim_{\hbar \rightarrow 0} \langle \eta(t) \nu'(t') \rangle_r = \langle \eta_{\text{cl}}(t) \nu'_{\text{cl}}(t') \rangle_r = 0. \quad (3.126)$$

Since the ν' noise is now entirely uncorrelated, it will have no effect on the average dynamics and can be dropped from the action. This restores symmetry to the forwards and backwards propagations

$$\tilde{S}_{\text{cl}} = \int_0^{t_f} dt L_Q(q, t) - \eta(t) f(q, t). \quad (3.127)$$

The classical equation of motion we obtain for a single trajectory is therefore a type of Langevin equation:

$$\ddot{q} = -\frac{\partial V(q)}{\partial q} + \eta_{\text{cl}}(t) f'(q, t) \quad (3.128)$$

It is not a surprise that the classical limit of the ESLE corresponds to a Langevin equation, but Eq. (3.128) appears to lack some essential features. Most concerningly, the ν' noise appears to have no effect on the dynamics. This is a consequence of incorporating the dynamic response of the bath into the HS transformation, and appears to be irretrievably lost in the classical limit. There is also the question of what the imaginary time noise corresponds to in the classical limit. Since position and momentum now commute, calculating the thermal reduced probability density classically is trivial, and the need to describe the initial density as a path integral is obviated. While the imaginary noise retains non-zero correlations, they are entirely

ancillary to the classical system dynamics.

Neither of these issues can be satisfactorily answered here. Taking limits is a messy business and while we can sketch the essential behaviour of the classical system, there are some subtle issues being passed over without comment. To address this, chapter 5 will construct the KvN path integral and perform an equivalent analysis of the CL model. In the process, we will establish the correct classical equivalent to the ESLE and resolve the issue of both the ν and $\bar{\mu}$ noises.

Aside: Interpreting the Trajectories

We have found the classical limit of the ESLE is a Langevin equation, featuring the same stochastic trajectories as the quantum case. The stochastic trajectories arose as a formal device in the HS transformation for describing an effective Hamiltonian, without reference to the physical content of this transformation. The interpretation of stochastic terms in the classical context is straightforward [69], but what about in the fully quantum case? Is it possible to attach a physical interpretation to an individual trajectory?

Critically, for an element of a formalism to be physically meaningful, it must be possible to isolate its effect on observations. In the purely classical case this is not a problem, as any given trajectory has an associated probability to be observed, entirely dependent on the initial condition of the combined system and bath³.

The quantum case is no different, in the sense that the stochastic terms are also capturing the effect of an unknown initial state sampled from some probability distribution. The difference lies in the fact that in the quantum regime the statistical distribution of an observable is due to both the probabilistic sampling of an initial

³This line of thinking has proven to be extraordinarily productive, spawning the field of stochastic thermodynamics. In particular, it is responsible for the Jarzynski/Crooks fluctuation-dissipation relations, perhaps the only genuinely *new* result in thermodynamics for the past century.

state, *and* the inherently quantum nature of the system evolution. In order to draw out a physical interpretation, it must be possible to disentangle these two contributions, such that one can identify the ensemble of realisations generated by the *same* initial state.

Explicitly, let us consider an observable expectation A for the Q subsystem. The expectation of such an observable will be given by

$$\langle \hat{A}(t) \rangle = \text{Tr} \left(\langle \tilde{\rho}(t) \rangle_r \hat{A} \right) = \int dq A(q, t). \quad (3.129)$$

As both the trace operation and the stochastic averaging are linear operations, we can swap the order of averaging

$$\langle \hat{A}(t) \rangle = \left\langle \text{Tr} \left(\tilde{\rho}(t) \hat{A} \right) \right\rangle_r = \langle A_j(t) \rangle_r. \quad (3.130)$$

The object $A_j(t)$ is the ensemble average for a single noise trajectory, labeled by j . If the system-environment was initialised in a definite state (rather than a statistical mixture of states), we would always obtain some $A_j(t)$ as the expectation for \hat{A} , with different initial states corresponding to different trajectories. Furthermore, this single-trajectory expectation may be expressed as the ensemble average of $A_j(q, t)$:

$$A_j(t) = \int dq A_j(q, t) \quad (3.131)$$

leading to the interpretation of $A_j(q, t)$ as an observable value found from a single measurement. The integration over its dependent coordinate is the ensemble average, while averaging over the j index performs the stochastic average.

The two different processes are illustrated in Fig. 3.3, demonstrating the way one may partition the overall expectation into averages over either the initial state or the ensemble, with the full observable expectation being recovered when averaging over

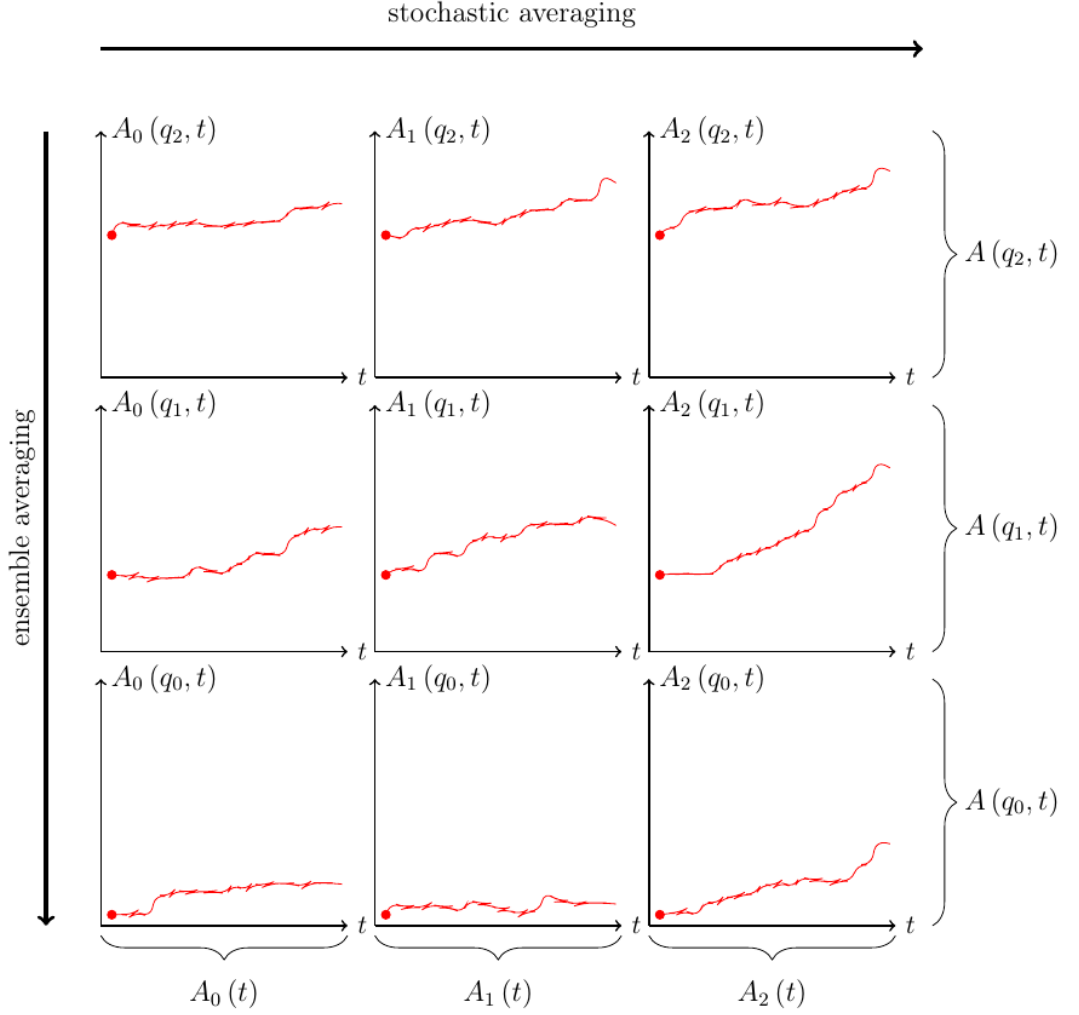


Figure 3.3: Schematic demonstrating the full ensemble over different initial system+bath states. The braces indicate the result when averaging over the appropriate dimension. Averaging over both the ensemble and stochastic trajectory would yield the experimentally observed expectation $\langle \hat{A}(t) \rangle = A(t)$.

both. In this sense it is possible to assign an interpretation to a given stochastic trajectory, i.e. that its expectations are what we would observe for a system evolving from a given pure state for the composite system.

If one were to clone a given initial state many times and perform statistics on its evolution, then $A_j(t)$ would be directly observable and undeniably physical. Unfortunately, the quantum no-cloning theorem [138] explicitly prohibits the copying of an arbitrary state, rendering the notion of ensemble averaging over a single trajectory unrealisable. All of this is to say that the effect of the initial state cannot be isolated and $\tilde{A}(q, t)$ is *impossible* to access experimentally. In a strictly statistical interpretation of quantum mechanics, a single stochastic realisation has *no* physical significance⁴ and the ESLE only acquires meaning after performing the stochastic averaging. In the classical limit, the ensemble averaging process becomes redundant (in the context of a system initialised in a definite state) and hence an unproblematic interpretation of the trajectories is recovered.

3.5.3 Hamiltonian Of Mean Force

The ESLE is a composition of two differential equations, but yields useful results even for when the total system is in equilibrium. In this case the imaginary time differential equation allows for the exact calculation of the *reduced equilibrium density matrix*. This is important, as the stationary distribution of dissipative systems with finite couplings has been shown to deviate from that expected under partitioned conditions [139]. Describing the statistics of such a system is not trivial, necessitating a formulation of thermodynamic laws beyond the assumption of weak environmental coupling [140]. This is sometimes achieved by defining a “Hamiltonian of mean force” \hat{H}_{MF} . This is the effective Hamiltonian that describes the canonical distribution of

⁴Assigning meaning to a single trajectory falls into the same category of lunacy as divining the nature of wavefunction collapse, i.e. trying to infer from a statistical theory something tangible about a single measurement.

the open system after tracing out the bath:

$$\hat{H}_{MF} = -\frac{1}{\beta} \ln \left(\frac{\text{Tr}_X \left[e^{-\beta \hat{H}_{\text{tot}}} \right]}{\text{Tr} \left[e^{-\beta \hat{H}_{\text{tot}}} \right]} \right) \quad (3.132)$$

such that the reduced system in equilibrium is described by

$$\rho_0 = \frac{1}{Z} e^{-\beta \hat{H}_{MF}}. \quad (3.133)$$

Clearly \hat{H}_{MF} is closely related to the reduced canonical density given in Eq.(3.107). Using this, it is possible to approximate the Hamiltonian of mean force for a CL environment. To do so we start from the simplified model

$$\tilde{\rho}_0 = \hat{\tau} \exp \left[-\beta \hat{H}_Q - \frac{1}{\hbar} \hat{f} \int_0^{\beta \hbar} d\tau \bar{\mu}(\tau) \right]. \quad (3.134)$$

Only the noise $\bar{\mu}$ is dependent on imaginary time, so for the sake of concision we have dropped time labels for operators and will reinsert them into the final result. Upon averaging, this will give the physical initial reduced density matrix in Eq. (3.133) and hence a way to assign \hat{H}_{MF} . As a first approximation we express $\tilde{\rho}_0$ as

$$\tilde{\rho}_0 \approx e^{-\beta \hat{H}_Q} \hat{\tau} \exp \left[-\frac{1}{\hbar} \hat{f} \int_0^{\beta \hbar} d\tau \bar{\mu}(\tau) \right]. \quad (3.135)$$

The obvious problem here is the uncontrolled approximation⁵ when $[\hat{H}_Q, \hat{f}]_- \neq 0$. Despite this, we are at least able to draw out some important characteristics of the Hamiltonian of mean force, namely that it depends on both temperature and the *square* of the system-environment coupling.

⁵Due to $\bar{\mu}$ averaging to zero, this approximation yields the same results as Taylor expanding the full exponential to second order.

To evaluate the average, we first expand the stochastic exponential

$$\hat{\tau} \exp \left[-\frac{1}{\hbar} \hat{f} \int_0^{\beta \hbar} d\tau \bar{\mu}(\tau) \right] = \sum_n \frac{1}{n!} \left(-\frac{1}{\hbar} \hat{f} \int_0^{\beta \hbar} d\tau \bar{\mu}(\tau) \right)^n. \quad (3.136)$$

Any term with an odd power of \hat{f} in this expansion can be discarded, since from Isserlis'/Wick's theorem [141] the average over an odd product of zero-mean Gaussian noises is zero:

$$\left\langle \exp \left[-\frac{1}{\hbar} \hat{f} \int_0^{\beta \hbar} d\tau \bar{\mu}(\tau) \right] \right\rangle_{\bar{z}(\tau)} = \sum_n \frac{1}{(2n)!} \left\langle \left(-\frac{1}{\hbar} \hat{f} \int_0^{\beta \hbar} d\tau \bar{\mu}(\tau) \right)^{2n} \right\rangle_r. \quad (3.137)$$

Wick's theorem can also be used to assess this average when n is even

$$\left\langle \left(\int d\tau \bar{\mu}(\tau) \right)^{2n} \right\rangle_r = \frac{(2n)!}{2^n n!} C(\beta)^n \quad (3.138)$$

$$C(\beta) = \int d\tau d\tau' \langle \bar{\mu}(\tau) \bar{\mu}(\tau') \rangle_r. \quad (3.139)$$

Note that $C(\beta)$ is completely specified when $I(\omega)$ is provided. Substituting this into the series expansion yields:

$$\sum_n \frac{1}{(2n)!} \left\langle \left(-\frac{1}{\hbar} \hat{f} \int_0^{\beta \hbar} d\tau \bar{\mu}(\tau) \right)^{2n} \right\rangle_r = \sum_n \frac{1}{2^n n!} \left(-\frac{1}{\hbar} \hat{f} \right)^{2n} C(\beta)^n \quad (3.140)$$

$$\Rightarrow \left\langle \exp \left[-\frac{1}{\hbar} \hat{f} \int_0^{\beta \hbar} d\tau \bar{\mu}(\tau) \right] \right\rangle_r = \exp \left[\frac{1}{2\hbar^2} \hat{f}^2 C(\beta) \right] \quad (3.141)$$

which can be used to give a first approximation to the Hamiltonian of mean force for CL models

$$\hat{H}_{MF} \approx \hat{H}_Q - \frac{1}{2\beta \hbar^2} \hat{f}^2 C(\beta). \quad (3.142)$$

As promised, this approximation elucidates the essential temperature and coupling

dependence of the mean force Hamiltonian. This is of course contingent on the approximation being reasonable and while it would be nice to do away with it entirely, I believe doing so would require a mastery of combinatorial properties that is simply beyond me.

3.5.4 Attaching A Second Bath

Finally, we can consider the effect of connecting our open system to a second reservoir. In general, this would produce an additional set of noises such that the real-time part of the ESLE would now read

$$\begin{aligned}
i\hbar\partial_t\tilde{\rho}(t) = & \left[\hat{H}_Q(t), \tilde{\rho}(t)\right]_- - \eta_1(t) \left[\hat{f}_1(t), \tilde{\rho}(t)\right] - \frac{\hbar}{2}\nu_1(t) \left\{\hat{f}_1(t), \tilde{\rho}(t)\right\} \\
& - \eta_2(t) \left[\hat{f}_2(t), \tilde{\rho}(t)\right] - \frac{\hbar}{2}\nu_2(t) \left\{\hat{f}_2(t), \tilde{\rho}(t)\right\}
\end{aligned} \tag{3.143}$$

where the subscripts indicate which environment a noise and coupling belongs to.

For the environments to be distinguishable, they should not interact with each other (or they would equilibrate), so the set of noises associated with each bath will be uncorrelated to each other. If the two baths are at different temperatures, the total system cannot initially be in thermal equilibrium. The initial condition can only support one bath in equilibrium with the open system, and the imaginary time evolution is identical to that without the second (initially decoupled) environment. The imaginary and cross-time correlations will therefore be associated with only one bath.

In some special cases it is possible to reduce Eq.(3.143) to an effective single environment description. To do so, we require that the power spectrum and system-bath coupling for both environments is identical, but allow them separate inverse temperatures β_1 and β_2 . We can describe the real time evolution with Eq.(3.114) with the following redefinition of the noises:

$$\eta(t) = \eta_1(t) + \eta_2(t) \quad (3.144)$$

$$\nu(t) = \nu_1(t) + \nu_2(t) \quad (3.145)$$

The subscripted noises have the correlation functions described by Eqs. (3.115-3.119). The cross-correlation between these two noises is a simple scaling:

$$\langle \eta(t) \nu(t') \rangle_r = 2K_{\eta\nu}(t - t') \quad (3.146)$$

while the auto-correlation of η has a *new* functional form

$$\begin{aligned} \langle \eta(t) \eta(t') \rangle_r &= \langle \eta_1(t) \eta_1(t') \rangle_r + \langle \eta_2(t) \eta_2(t') \rangle_r \\ &= \hbar \int_0^\infty \frac{d\omega}{\pi} I(\omega) \left[\coth\left(\frac{1}{2}\omega\hbar\beta_1\right) + \coth\left(\frac{1}{2}\omega\hbar\beta_2\right) \right] \cos(\omega(t - t')). \end{aligned} \quad (3.147)$$

Using the compound angle hyperbolic identities, we can express this in a more compact way

$$\langle \eta(t) \eta(t') \rangle_r = 2\hbar \int_0^\infty \frac{d\omega}{\pi} I(\omega) R(\omega, \bar{\beta}, \Delta) \coth\left(\frac{1}{2}\omega\hbar\bar{\beta}\right) \cos(\omega(t - t')) \quad (3.148)$$

$$R(\omega, \bar{\beta}, \Delta) = \frac{\sinh^2\left(\frac{1}{2}\bar{\beta}\hbar\omega\right)}{\sinh^2\left(\frac{1}{2}\bar{\beta}\hbar\omega\right) - \sinh^2\left(\frac{1}{2}\Delta\hbar\omega\right)} \quad (3.149)$$

where $\bar{\beta} = \frac{\beta_1 + \beta_2}{2}$ is the average inverse temperature and $\Delta = \beta_1 - \beta_2$ is the difference.

When $\frac{\bar{\beta}}{\Delta} \gg 1$, $R(\omega, \bar{\beta}, \Delta) \approx 1$ and in this case the correlation function is also a simple scaling of Eq. (3.115). It is then possible to describe the effect of a second environment as a modified single bath at $\bar{\beta}$ and spectral density $I(\omega) \rightarrow 2I(\omega)$ (i.e. an effective doubling of the coupling strength between system and bath). Beyond this limit, the non-trivial functional form of R demonstrates the impossibility of capturing

the effect of two baths with an appropriate parametrisation of a single bath model.

3.6 Chapter Summary

The fundamental result of this chapter is the removal of the partitioned initial condition which implied that the open system and the bath were initially isolated. The form of the model interaction has also been generalised, but is still limited by the essential need for an interaction to be linear in environmental oscillator displacements. This generalised Hamiltonian emerges naturally from an arbitrary total system Hamiltonian by expanding atomic displacements of the environment up to the second order, and can therefore be directly applied to approximate more realistic systems.

Following procedures to accommodate a more physical partition-free approach, we applied a special variant of the HS transformation that allowed the initial condition to be determined via an auxiliary differential equation. This allows the ESLE to make exact predictions for the transient behaviour of the primary system when it is perturbed from equilibrium.

The ESLE represents a unification and generalisation of the differential equations derived by Stockburger [85] and Moix *et al.* [142], resulting in additional and highly non-trivial constraints on the correlations between the real and imaginary time noises. The connection between these two pieces of work was not previously apparent, but has emerged naturally from the simultaneous generalisation of the model Hamiltonian and the initial total density matrix. This is the ESLE's principal advantage, and allows for a simpler and more general closed form description of the evolution of the reduced density matrix, as compared to hierarchical equations of motion [143].

The ESLE also contains a potential route to generating the calculation of a broad class of Hamiltonians of mean force, but to do so exactly will require a more sophisticated derivation than was sketched in the previous section. We also note that

this approach can easily be generalised to several environments, e.g. for heat transport problems along similar lines to Ref. [89]. This does however necessitate either a redefinition of the noise kernels, or the addition of extra noises.

Extracting numerical results from the ESLE depends on the feasibility of generating noises that satisfy the correlations outlined in section 3.3. Implementing this for a simple two-state system will be the focus of the next chapter.

Chapter 4

Numerical Implementation

Computers are useless. They can
only give you answers.

Pablo Picasso

After deriving the ESLE, it is natural to ask whether it is possible to perform practical computations using it. In this chapter we present the first application of the ESLE method to a calculation *in silico*, originally published in Ref. [144].

Our aim here is (i) to introduce a numerical implementation of the ESLE, and (ii) apply this formalism to a spin-boson system driven from equilibrium. Particular attention is paid to investigating the evolution of this model system under a Landau-Zener sweep. We show that the ESLE predicts differences in both the short- and long-time reduced density matrix evolution, including the asymptotic regime, as compared to partitioned approaches, in which various initial density matrices are assumed. This demonstrates the importance of the correct initial preparation of the system, manifested by the correlations between the imaginary and real time evolutions of the open system.

The first part of this chapter, section 4.1, consists of an exposition on the methods used to generate the noise terms. Section 4.2 presents the spin-boson Hamiltonian,

while section 4.3 details the results of driven simulations using the ESLE. We close with a discussion on the range of applications and the limitations of the current numerical implementation of ELSE in section 4.4.

4.1 Generating Noises

The numerical scheme for the ESLE is in principle rather simple. The procedure is to generate a realisation of the noises satisfying Eqs. (3.115)-(3.118), evolve the density matrix according to Eqs. (3.113) and (3.114), and finally average over realisations. In this section we shall discuss how for the simplified model detailed in section 3.5.1 noises are generated; the method used can then easily be generalised to any number of noises.

The general scheme to generate coloured Gaussian noises is well known [145], however the ESLE requires the generation of noises with cross-correlations (in two different time dimensions), which obey the specific statistical relationships detailed in section 3.5.1. This requires a slight generalisation of the usual methods.

In order to generate the noise, we begin by splitting each noise into sub-terms which are only correlated with one other term across the noises:

$$\eta(t) = \eta_\eta(t) + \eta_\nu(t) + \eta_{\bar{\mu}}(t) \quad (4.1)$$

$$\nu(t) = \nu_\eta(t) \quad (4.2)$$

$$\bar{\mu}(t) = \bar{\mu}_{\bar{\mu}}(\tau) + \bar{\mu}_\eta(\tau) \quad (4.3)$$

where (for example), the η_η noise is the auto-correlative part of the total η noise, and has a non-zero correlation only with itself, while the $\eta_{\bar{\mu}}$ noise only correlates with the

$\bar{\mu}_\eta$ term. To enforce this effective “noise-orthogonality”, we express each term as a convolution between a filtering kernel (all such kernels are denoted as $G_{\eta\eta}$, $G_{\eta\nu}$, etc.) and one of a number of real white noise processes $\{x_i(t)\}$ and $\{\bar{x}_i(\tau)\}$, which have the property:

$$\langle x_i(t) x_j(t') \rangle_r = \delta_{ij} \delta(t - t') \quad (4.4)$$

$$\langle \bar{x}_i(\tau) \bar{x}_j(\tau') \rangle_r = \delta_{ij} \delta(\tau - \tau') \quad (4.5)$$

$$\langle x_i(t) \bar{x}_j(\tau) \rangle_r = 0. \quad (4.6)$$

Given that the time (real or imaginary) is simply a parameter in the noise process, the distinction between x_i and \bar{x}_j is one of notational convenience, rather than an expression of any fundamentally dissimilar statistics. The various components of the three complex noises we need are generated by the following convolutions of the filtering kernels with the white noises:

$$\eta_\eta(t) = \int_{-\infty}^{\infty} dt' G_{\eta\eta}(t - t') x_1(t') \quad (4.7)$$

$$\eta_\nu(t) = \int_{-\infty}^{\infty} dt' G_{\eta\nu}(t - t') (x_2(t') + ix_3(t')) \quad (4.8)$$

$$\nu_\eta(t) = \int_{-\infty}^{\infty} dt' G_{\nu\eta}(t - t') (x_3(t') + ix_2(t')) \quad (4.9)$$

$$\bar{\mu}_{\bar{\mu}}(\tau) = \int_{-\beta\hbar}^{\beta\hbar} d\tau' G_{\bar{\mu}\bar{\mu}}(\tau - \tau') \bar{x}_1(\tau') \quad (4.10)$$

$$\eta_{\bar{\mu}}(t) = \int_0^{\beta\hbar} d\tau' G_{\eta\bar{\mu}}(t, \tau') (\bar{x}_2(\tau') + i\bar{x}_3(\tau')) \quad (4.11)$$

$$\bar{\mu}_{\eta}(\tau) = \int_0^{\beta\hbar} d\tau' G_{\bar{\mu}\eta}(\tau, \tau') (\bar{x}_3(\tau') + i\bar{x}_2(\tau')). \quad (4.12)$$

Here the limits on integrations over imaginary time reflect the fact that τ is constrained to lie within the interval $[0, \beta\hbar]$.

In this construction the various filtering kernels are yet to be determined. Importantly, as the only physically relevant constraints on the noises are the physical kernels K (see Eqs. (3.115) - (3.119)), this linear filtering ansatz will be valid provided we can establish a consistent mapping between the physical K and filtering G kernels. Note that for correlations in the same time dimension we assume the filtering kernels G have the same stationarity properties (i.e. they depend only on time differences) as the corresponding physical kernels K . The *cross-time* filtering kernels $G_{\eta\bar{\mu}}(t, \tau)$ and $G_{\bar{\mu}\eta}(\tau, \tau')$, that are responsible for the *cross-time* correlation $\langle \eta(t) \bar{\mu}(\tau) \rangle_r$ between real and imaginary time noises, are not assumed to be stationary.

It can easily be seen that with the above choice, $\langle \eta(t) \eta(t') \rangle_r = \langle \eta_{\eta}(t) \eta_{\eta}(t') \rangle_r$, $\langle \nu(t) \eta(t') \rangle_r = \langle \nu_{\eta}(t) \eta_{\nu}(t') \rangle_r$, $\langle \bar{\mu}(\tau) \bar{\mu}(\tau') \rangle_r = \langle \bar{\mu}_{\bar{\mu}}(\tau) \bar{\mu}_{\bar{\mu}}(\tau') \rangle_r$, and $\langle \eta(t) \bar{\mu}(\tau) \rangle_r = \langle \eta_{\bar{\mu}}(t) \bar{\mu}_{\eta}(\tau) \rangle_r$. All other correlation functions are identically equal to zero because of the design imposed “orthonormality” of the white noises, Eqs. (4.4)-(4.6). For instance,

$$\begin{aligned} \langle \nu(t) \nu(t') \rangle_r &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dt_1 dt_2 G_{\nu\eta}(t - t_1) G_{\nu\eta}(t' - t_2) \\ &\times (\langle x_3(t_1) x_3(t_2) \rangle_r - \langle x_2(t_1) x_2(t_2) \rangle_r + 2i \langle x_3(t_1) x_2(t_2) \rangle_r) = 0 \end{aligned} \quad (4.13)$$

where in the last equality we have applied Eq. (4.4).

To find the correspondence between the physical and filtering kernels, we substitute the assumed functional form of each noise given above into Eqs. (3.115)-(3.118).

Explicit evaluation of the η auto-correlative component yields:

$$\begin{aligned}\langle \eta(t) \eta(t') \rangle_r &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dt_1 dt_2 G_{\eta\eta}(t-t_1) G_{\eta\eta}(t'-t_2) \langle x_1(t_1) x_1(t_2) \rangle \\ &= \int_{-\infty}^{\infty} dt_1 G_{\eta\eta}(t-t_1) G_{\eta\eta}(t'-t_1)\end{aligned}\quad (4.14)$$

leading to the first of the kernel mappings:

$$K_{\eta\eta}(t-t') = \int_{-\infty}^{\infty} dt_1 G_{\eta\eta}(t-t_1) G_{\eta\eta}(t'-t_1). \quad (4.15)$$

The physical kernel is therefore expressible as a self-convolution of the filtering kernel, and can be further simplified using its Fourier representation

$$\tilde{K}_{\eta\eta}(\omega) = \left| \tilde{G}_{\eta\eta}(\omega) \right|^2. \quad (4.16)$$

This equation is the *only* constraint on $G_{\eta\eta}$, and any solution to this equation yields a valid filtering kernel. In this particular case the solution is unique (up to a phase), but we shall see later that cross-correlative mappings do *not* uniquely define the relevant filtering kernels, and hence some choice exists which can be exploited. Given that the physical kernel here is both real and symmetric, we may constrain the filtering kernel to have the same properties and express it simply as:

$$\tilde{G}_{\eta\eta}(\omega) = \sqrt{\tilde{K}_{\eta\eta}(\omega)} \quad (4.17)$$

The auto-correlative component of the $\bar{\mu}$ noise has the same properties as above, provided we extend the integration domain of Eq. (4.10) and periodically extend $G_{\bar{\mu}\bar{\mu}}(\tau - \tau')$ across this domain. Then the filtering kernel has an identical mapping in Fourier space:

$$\tilde{G}_{\bar{\mu}\bar{\mu}}(\omega) = \sqrt{\tilde{K}_{\bar{\mu}\bar{\mu}}(\omega)} \quad (4.18)$$

There are also two non-zero cross-correlative terms to consider: the real time correlation between the η and ν noises, and the cross-time correlation between η and $\bar{\mu}$. In the first case, we have:

$$\langle \eta(t) \nu(t') \rangle_r = K_{\eta\nu}(t - t') = 2i \int_{-\infty}^{\infty} dt_1 G_{\eta\nu}(t - t_1) G_{\nu\eta}(t' - t_1) \quad (4.19)$$

or in Fourier space:

$$\tilde{K}_{\eta\nu}(\omega) = 2i \tilde{G}_{\eta\nu}(\omega) \tilde{G}_{\nu\eta}^*(\omega) \quad (4.20)$$

Unlike with the auto-correlative processes, we are left with a degree of choice in the form of the two filtering kernels. Here we take the simple expedient of choosing one of the kernels as a delta function, $G_{\nu\eta}(t) = \delta(t)$ or $\tilde{G}_{\nu\eta}(\omega) = 1$. The second filtering kernel may therefore be straightforwardly identified as:

$$\tilde{G}_{\eta\nu}(\omega) = -\frac{i}{2} \tilde{K}_{\eta\nu}(\omega) \quad (4.21)$$

We now turn our attention to the cross-time correlation $K_{\eta\bar{\mu}}(t - i\tau)$, for which we obtain:

$$\langle \eta(t) \bar{\mu}(\tau) \rangle_r = K_{\eta\bar{\mu}}(t - i\tau) = 2i \int_0^{\beta\hbar} d\tau' G_{\eta\bar{\mu}}(t, \tau') G_{\bar{\mu}\eta}(\tau, \tau') \quad (4.22)$$

In this case we cannot use the Fourier transformation to simplify the expression of the filtering kernels, as the physical kernel is inherently non-stationary. Once again it is convenient to set one kernel as a delta function, $G_{\bar{\mu}\eta}(\tau, \tau') = \delta(\tau - \tau')$, giving the form of the other kernel as:

$$K_{\eta\bar{\mu}}(t - i\tau) = 2iG_{\eta\bar{\mu}}(t, \tau) \quad (4.23)$$

which completes the mapping between the physical and filtering kernels. Note that setting either of the filtering kernels $G_{\eta\bar{\mu}}(t, \tau)$ or $G_{\bar{\mu}\eta}(\tau, \tau')$ to zero results in the loss of correlations between the imaginary and real time evolutions. This corresponds to the Stochastic Liouville-von Neumann Equation (SLE) method [85].

Armed with this mapping, the noises are straightforwardly generated by the convolution of filtering kernels with vectors of white noise (with variances appropriate to the discretisation of the chosen time step).

4.1.1 Numerical Implementation Algorithm

The process for simulating a single trajectory for the ESLE consists of two parts: generating noise vectors, and using those in a stochastic differential equation. When generating noises, we have seen that each noise is a sum of different components with constructed correlations. In general the form of these noise components when discretised is:

$$y_i = \delta_{t/\tau} \sum_j g_{ij} x_j \quad (4.24)$$

where δ_t (or δ_τ) is the step for the real (imaginary) time, x is a complex sum of unit variance white noises scaled by $1/\sqrt{\delta_{t/\tau}}$ (to give the discrete delta function correlations), and g_{ij} is the (discretised) appropriate filtering kernel. The second index on g is necessary for describing the cross-time correlative components of the $\bar{\mu}$ and η noises, where the filtering kernel is inherently two-dimensional. In this case Eq. (4.24) must be implemented through direct matrix multiplication. For example, $\eta_{\bar{\mu}}$ is calculated as:

$$\eta_{\bar{\mu}}(t_i) = \delta_{\tau} \sum_{j=0}^M G_{\eta_{\bar{\mu}}}(t_i, \tau_j) (\bar{x}_2(\tau_j) + i\bar{x}_3(\tau_j)) \quad (4.25)$$

where $i = 1, \dots, N$ ($j = 1, \dots, M$), with N being the number of real timesteps (M imaginary timesteps). This method is expensive, as it requires $N \times M$ operations to generate the cross-time component of a noise vector.

For components of the noise with stationary correlations (i.e. no mixing of real or imaginary time), the filtering kernel matrix is expressible as a vector of time differences, $g_{ij} \rightarrow g_{i-j}$. It is therefore much more efficient to use the Fast Fourier Transform (FFT) to convolve the filtering kernels with the white noises. The FFT uses the Discrete Fourier Transform (DFT), which transforms a length N sequence in the following manner:

$$\text{DFT}_{\alpha}[\mathbf{y}] = Y_{\alpha} = \sum_{j=0}^{N-1} y_j \exp\left(-2\pi i \frac{\alpha j}{N}\right) \quad (4.26)$$

with the inverse:

$$\text{DFT}_j^{-1}[\mathbf{Y}] = y_j = \frac{1}{N} \sum_{\alpha=0}^{N-1} Y_{\alpha} \exp\left(2\pi i \frac{\alpha j}{N}\right) \quad (4.27)$$

The DFT has a *circular* convolution theorem:

$$\sum_{k=0}^{N-1} (g^{(N)})_{j-k} x_k = \text{DFT}_j^{-1} \left[\sum_{\alpha} G_{\alpha} X_{\alpha} \right] \quad (4.28)$$

where $g^{(N)}$ is the periodic extension of the sequence g :

$$(g^{(N)})_i \equiv g_{i \pmod{N}} \quad (4.29)$$

It is possible to obtain the linear convolution from the circular convolution (and hence the DFT convolution theorem) by padding both the filtering kernel and noise vectors

with a large number of zeros [146]. This allows an efficient generation of stationary noise components as:

$$y_j = \delta_{t/\tau} \text{DFT}_j^{-1} \left[\sum_{\alpha} \text{DFT}_{\alpha} [\mathbf{g}] \text{DFT}_{\alpha} [\mathbf{x}] \right] \quad (4.30)$$

Typical results for noise generation are shown in Fig. 4.1. The auto-correlation of η and its cross-correlation with ν are shown in a) and b) respectively, while the cross-time correlations c), d) and e), f) are illustrated with two separate τ slices. In many cases the generated noise correlation and the analytic kernel are close enough to overlap on the figure. Correlations expected to be zero are not shown, but the *maximum* value found across any of these functions was on the order of 10^{-4} . The final figure h) shows the RMS deviation between the (real part) of the generated noise covariances and their respective kernels with increasing run number.

This figure demonstrates that excellent convergence to the physical kernels can be achieved, provided that a sufficient sampling is taken¹. In particular, the apparently noisier behaviour of $\langle \bar{\mu}(\tau) \bar{\mu}(\tau') \rangle_r$ is due to its relatively small range of values, and the fact that its cross-time correlative part must be generated with the cruder direct summation.

¹For correlations in the same time dimension, this is actually only on the order of 10^3 , but including cross-time correlations requires around 10^5 samples, depending on coupling strength.

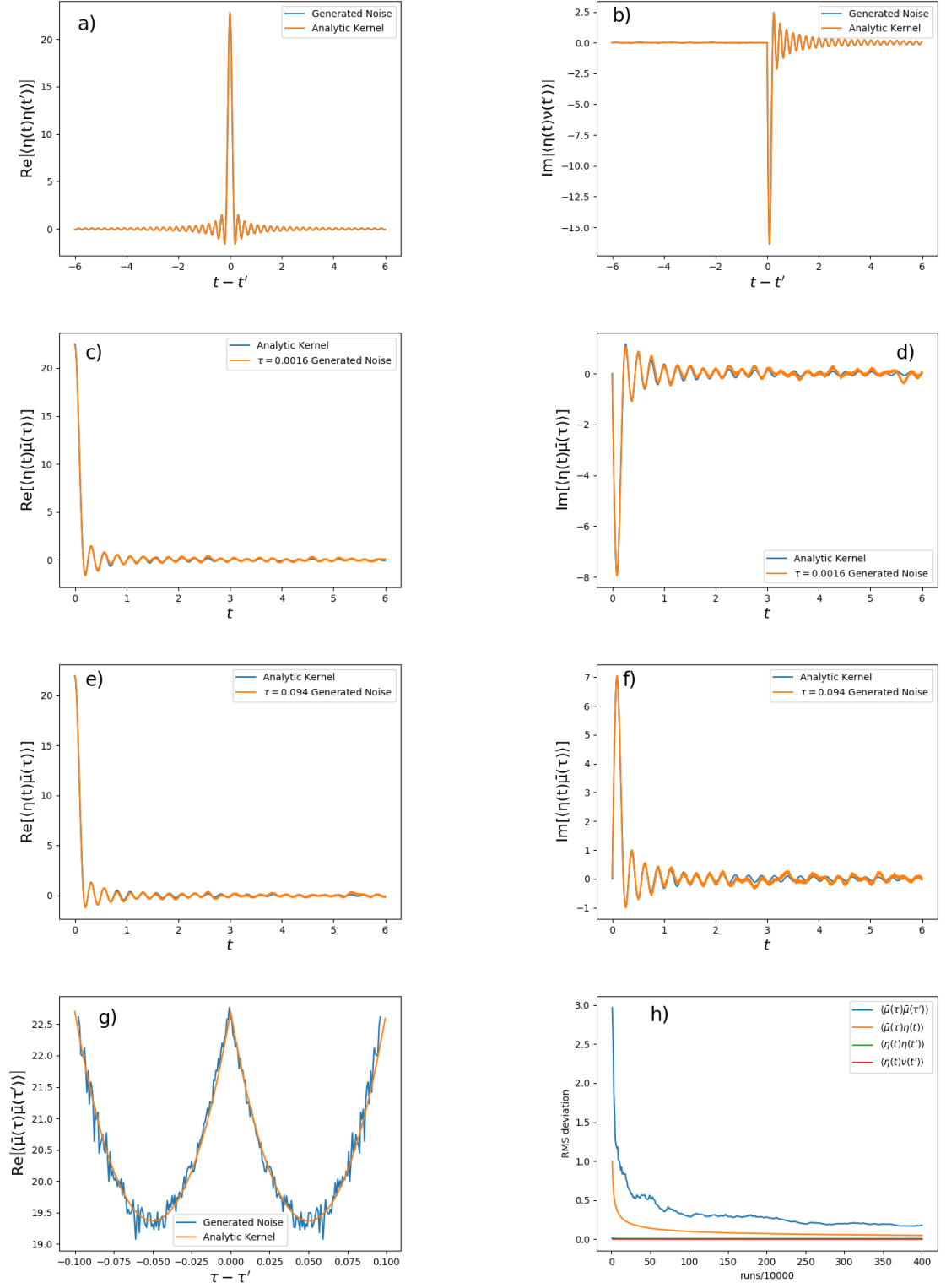


Figure 4.1: Typical correlation functions obtained from generated noise after 4×10^5 runs with parameters $\beta = 0.1\Delta^{-1}$, $\alpha = 0.2$, $\omega_c = 25$ and an Ohmic spectral density.

4.2 spin-boson Models

Having successfully generated noises which obey the appropriate correlation functions, we now turn our attention to applying the ESLE to a specific system. In this regard, the spin-boson model is a particularly popular test-bed [12, 82, 83, 91, 147, 148]. This model has also been interrogated by other methods, including cumulant expansions [149], matrix products [150] and reaction-coordinate [63] approaches. It is also amenable to analytic derivations, both perturbatively [151] and nonperturbatively [152], as well as to an application of the Born approximation [153]. The model may also draw on the extensive work done on driven two-state models, most famously the Landau-Zener sweep [154–156] and its generalisations [157, 158].

In addition, the spin-boson model displays rich, non-trivial behaviour, with integrable and non-integrable regimes [159], diffusive and localised phases [160], as well as coherent to incoherent crossovers [161, 162]. Besides the model’s obvious application to qubit behaviour, it has been mapped to impurities in an electronic bath (i.e. Kondo model) [163, 164], Josephson junctions [165–167], cold atoms [148, 168], and even biological systems [169]. The spin-boson model therefore serves as an excellent toy model, with application to real experimental systems.

4.2.1 Model Hamiltonian

The spin-boson model consists of a two-state system coupled to a CL environment. The two-state system is described by the (matrix) Hamiltonian:

$$H_q(t) = \epsilon(t) \sigma_z + \Delta(t) \sigma_x \quad (4.31)$$

Here $\epsilon(t)$ describes the bias between states, while $\Delta(t)$ controls tunneling between them; σ_z and σ_x are the usual Pauli spin-matrices. While using our formalism it would be possible to consider any spin-boson model, we shall focus here on two separate

protocols: (i) an equilibrium real time evolution when the bias $\epsilon(t)$ is kept constant at the value used to thermalise the whole system during imaginary time evolution, and (ii) a Landau-Zener type sweep, where the system has been thermalised at some time in the past, $t_0 < 0$, and then evolved with $\epsilon(t) = \kappa t$. In both cases the tunneling Δ is kept constant. As no analytic result exists for the second case, the asymptotic limit will be extrapolated numerically from the initial state calculated by the ESLE. Given that the ESLE evolves from an explicit thermal state at finite temperature and there is no analytic expression available, the two-state solution provides a useful numerical benchmark to evaluate the impact of the environment.

Coupling this two-state system to an environment of the simple CL type (see section 3.5.1) and using $f_\lambda = c_\lambda \sigma_z$, the total Hamiltonian reads

$$H_{\text{tot}}(t) = \epsilon(t)\sigma_z + \Delta\sigma_x + \sum_{\lambda} \hbar\omega_{\lambda} b_{\lambda}^{\dagger} b_{\lambda} - \sigma_z \sum_{\lambda} c_{\lambda} (b_{\lambda} + b_{\lambda}^{\dagger}). \quad (4.32)$$

This is simply the matrix form of the total Hamiltonian given in Eq. (1.22), with the appropriate model-specific substitutions. The environment operators have also been expressed using second quantisation, where b_{λ} (b_{λ}^{\dagger}) is the bosonic annihilation (creation) operator. The last term corresponds to the system-environment coupling which is proportional to the normal mode displacements of the environment. The only explicit t dependence in the total Hamiltonian is contained in the bias field for the open system.

To apply the ESLE to this system, we assume that the total system-environment is allowed initially (at time t_0) to thermalise having the Hamiltonian $H_0 = H_{\text{tot}}(t_0)$ corresponding to some initial value of the bias. Explicitly:

$$\rho_{\text{tot}}(t_0) = \frac{1}{Z_{\beta}} e^{-\beta H_0}. \quad (4.33)$$

This initial condition implies the following ESLE (matrix) equations in imaginary

($0 \leq \tau \leq \beta\hbar$) and real ($t \geq t_0$) times:

$$-\hbar\partial_\tau\bar{\rho}(\tau) = [\epsilon(t_0)\sigma_z + \Delta\sigma_x + \bar{\mu}(\tau)\sigma_z]\bar{\rho}(\tau) \quad (4.34)$$

$$i\hbar\partial_t\tilde{\rho}(t) = [\epsilon(t)\sigma_z + \Delta\sigma_x, \tilde{\rho}(t)]_- - \eta(t)[\sigma_z, \tilde{\rho}(t)]_- - \frac{\hbar}{2}\nu(t)\{\sigma_z, \tilde{\rho}(t)\} \quad (4.35)$$

In other words, we consider the initial condition to be parametrised by t_0 with real-time dynamics either keeping that value of the bias (the first protocol) or linearly driving the system away from its thermal state (the second).

Finally, as the ESLE does not begin from a partitioned state, it is an accurate description of the open system even in the strong-coupling regime. To parametrise the coupling strength and specify the environment model, we choose an Ohmic spectral density:

$$I(\omega) = \alpha\omega \left[1 + \left(\frac{\omega}{\omega_c} \right)^2 \right]^{-2} \quad (4.36)$$

where ω_c is some cut-off frequency. The parameter α is proportional to squares of the c_λ coefficients in Eq. (3.112) and hence describes the effective bath coupling strength. It has been shown that for $\alpha < \frac{1}{2}$ there is a coherent evolution, but crossing through the point $\alpha = \frac{1}{2}$ causes a phase change to incoherent spin dynamics [11, 170]. Beyond this at $\alpha > 1$ the system enters a localised regime where tunneling between the two states is completely suppressed (formally the bath coupling renormalises the tunneling element to $\Delta \rightarrow 0$). These behaviours are peculiar to the spin-boson model, and not indicative of a general restriction of the parameter space the ESLE is capable of simulating exactly. Our results will focus on the regime $\alpha < \frac{1}{2}$, where we should expect coherent, damped oscillations in the spin expectations.

4.2.2 Spin-boson Discretisation

In order to simulate the spin-boson model, we use a first order discretisation of the evolution equations:

$$\bar{\rho}_{\tau+\delta\tau} = \bar{\rho}_{\tau} + \delta\tau [\epsilon\sigma_z + \Delta\sigma_x + \bar{\mu}_{\tau}\sigma_z] \bar{\rho}_{\tau} \quad (4.37)$$

$$\tilde{\rho}_{t+\delta t} = \tilde{\rho}_t + i\hbar\delta t \left\{ [\epsilon\sigma_z + \Delta\sigma_x, \tilde{\rho}_t]_- - \eta_t [\sigma_z, \tilde{\rho}_t]_- - \frac{\hbar}{2}\nu_t [\sigma_z, \tilde{\rho}_t]_+ \right\} \quad (4.38)$$

This is essentially the Euler-Maruyama approximation. While the error of this discretisation is proportional to $\sqrt{\delta t/\tau}$, it is straightforward to implement directly, unlike more sophisticated schemes. Provided the timestep is small enough, it has proved sufficiently accurate for a first implementation of the ESLE (although this does require some convergence testing for a simulation over a given period). This is particularly important for the Landau-Zener sweep, where the frequencies of oscillations increase with time. An example of unphysical asymptotic behaviour with an insufficiently small timestep can be seen in Fig. 4.2.

4.2.3 Equilibrium Test

As a sanity check, we first test the ESLE with a time-independent bias. Given the ESLE is thermalised in imaginary time exactly, we expect the real-time evolution to show no change in the density matrix.

Figure 4.3 shows the density matrix components of both the ESLE, and a comparative simulation running the real time part of the ESLE without the cross-time correlations. This reduced case corresponds to the Stochastic Liouville Equation (SLE) [85] based on the partitioned approach. In the SLE case we initialise the density matrix

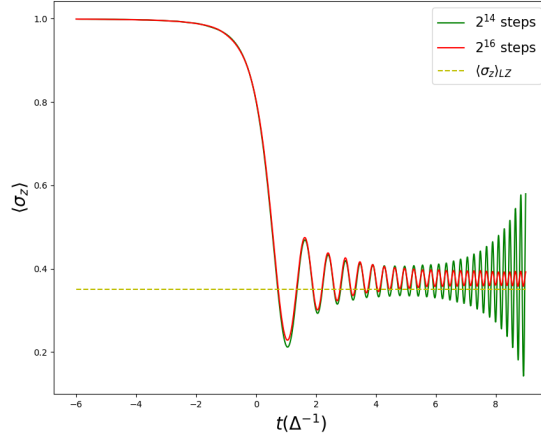


Figure 4.2: ESLE evolution for a fast sweep with $t_0 = -6\Delta^{-1}$, $\kappa = 4\Delta^2$, and $\alpha = 0.05$, sampled over 3×10^5 runs using 2^{14} and 2^{16} steps. The larger timestep (the green curve) leads to an unphysical magnification in the amplitude of oscillations (although the mean values are still comparable).

from two initial conditions: (i) $\rho_{ij} = \delta_{i1}\delta_{j1}$ and (ii) the density matrix predicted by the ESLE imaginary time evolution, $\rho_{ij} = \langle \bar{\rho}_{ij}(\hbar\beta) \rangle_r$.

For both the ESLE and the SLE simulation initialised from the imaginary-time evolution end-point, small oscillations in the components are observed, but they remain on average constant. This demonstrates that the cross-time correlations in the ESLE have little to no effect at equilibrium. This is as expected: any cross-time correlations in the noise rapidly die out as the system evolves in real time, and the noises evolving the SLE and ESLE become statistically identical. If the SLE simulation (started from the ESLE initial condition) evolved differently to the ESLE, then the ESLE would echo that behaviour later in time - a manifestly unphysical scenario at equilibrium.

The SLE simulation initialised at $\rho_{ij} = \delta_{i1}\delta_{j1}$, which represents a rather different density matrix to that expected at equilibrium, shows a relaxation of the spin. It is expected that this relaxation will converge to the same steady state as predicted

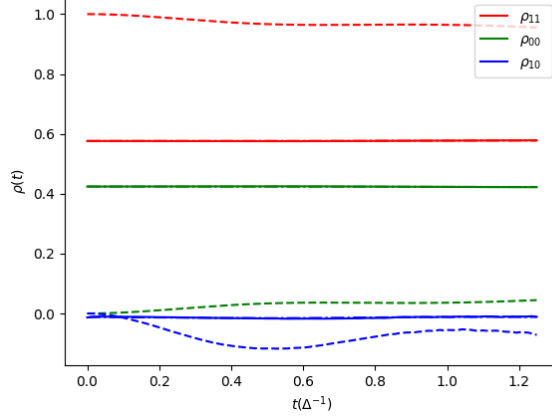


Figure 4.3: ESLE evolution of ρ for a time-independent system, averaged over 1×10^5 runs (solid lines). Dashed lines indicate an equivalent SLE evolution from $\rho_{ij} = \delta_{i1}\delta_{j1}$, while dash-dotted lines (not seen as they coincide with the solid lines) are SLE evolutions from $\rho_{ij} = \langle \bar{\rho}_{ij}(\hbar/\beta) \rangle_r$. Here $\Delta = 1$, $\epsilon = 5\Delta$, $\beta = 0.1\Delta^{-1}$, $\alpha = 0.05$, and $\omega_c = 200$. While the ESLE shows small fluctuations, the density matrix remains on average constant. The SLE evolution from $\rho_{ij} = \delta_{i1}\delta_{j1}$ predicts a relaxation to the ESLE values, but on a timescale not accessible by the simulation.

by the other simulations, but it does so on a timescale not fully accessible by our numerical scheme. This is due to the fact that in equilibrium, the cut-off frequency of the bath spectrum must be sufficiently large that any energy the spin system dissipates to the bath is returned in a finite time. If this is not the case thermal equilibrium is not possible, as the bath acts as an energy sink (causing the spin to relax). At the same time, from a numerical perspective, higher cut-off frequencies require a smaller time-step to avoid non-physical resonances². This is doubly problematic, as the timescale for the SLE to relax increases with cut-off frequency [171], while the stochastic simulation itself is step-limited. That is, numerical instabilities at longer times require excessive averaging to eliminate, i.e. many more simulations are needed for sampling, which is extremely demanding computationally. At lower cut-

²In reality, the cut-off frequency is fixed by the phonon lattice, but in simulations it is a useful parameter to have some control over.

off frequencies, all simulations are observed to converge to the same state, but the ESLE displays an unphysical spin-relaxation from the thermalised state due to the low cut-off.

4.3 Landau-Zener Sweep

Here we shall consider fully non-equilibrium simulations, in which the bias is linearly driven from the value $\epsilon(t_0) = \kappa t_0$ used for the equilibration (imaginary time evolution). This spin-boson model is particularly useful in this case, as for specific initial conditions the asymptotic behaviour can be analytically derived. In the zero temperature case, when the system is started in the state

$$\rho(-\infty) = \rho_0^{LZ} = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \quad (4.39)$$

and is decoupled from the environment oscillators, the asymptotic survival probability of that state is

$$P_{LZ} = \exp\left(-\frac{\pi\Delta^2}{\hbar\kappa}\right). \quad (4.40)$$

This protocol is known as the Landau-Zener (LZ) sweep, with P_{LZ} first derived by recasting the system as a Weber equation [154]. The result may also be found via contour integration [155] or direct evaluation of the time-ordered propagator [156]. This protocol has numerous experimental realisations, for example in Rydberg atoms [172] or Bose-Einstein condensates [173]. It has also been proven that the survival probability for the state is the same even with a σ_z coupling to a dissipative environment (of the Caldeira-Leggett type), with the caveat that the total system must be prepared in the ground state at zero temperature and evolved from $t_0 \rightarrow -\infty$ [174, 175]. Furthermore, numerical evaluations using a Stochastic Schrödinger Equation (SSE)

have shown that even in the case where the evolution is started from some finite time sufficiently far in the past, provided the bath coupling is weak ($\alpha < 0.2$), P_{LZ} is still recovered at zero temperature [82]. At stronger couplings however deviations from P_{LZ} in the asymptotic state were observed, confirming that generally even at zero temperature the asymptotic spin state in a dissipative system depends on both the bath coupling strength and the initial preparation of the system.

Given the ESLE is insensitive to the parameter values of the spin-boson system, it is interesting to explore the validity of the LZ sweep limit (4.40) in detail, both at finite temperature and with the environment coupling. Unlike in the equilibrium case, in order to simulate the driven system efficiently we must make an approximation to the ESLE. Principally this involves dropping the anticommutative term in Eq. (4.38) and using the effective evolution

$$i\hbar\partial_t\tilde{\rho}(t) = [\epsilon(t)\sigma_z + \Delta\sigma_x, \tilde{\rho}(t)]_- - \eta(t)[\sigma_z, \tilde{\rho}(t)]_- . \quad (4.41)$$

The numerical rationale for neglecting the ν term is due to its size in a typical trajectory as compared to the η noise. To see this, consider that the magnitude of the $G_{\eta\nu}$ kernel is much greater than that of the $G_{\eta\eta}$ filtering kernel (in Fourier space the cross correlation is proportional to the physical kernel, while the auto-correlative filtering kernel is equal to the *square root* of the physical kernel). Comparing the resultant η and ν noises when their respective filtering kernels are convolved with the same set of white noises, one finds that in natural units the ν noise is typically an order of magnitude greater than η . Because of this (except for a small window of parameters), a first order integrator, such as the one employed here, will fail to capture the correct behaviour of the system due to ν .

In order to understand the consequences of dropping the ν term, we must elucidate its physical interpretation. First, notice that this term pre-multiplies an anticommuta-

tor. Terms of this type are typically interpreted as the dissipative part of the equation of motion (as they are precisely the terms which break the time independence of the density matrix normalisation for individual trajectories). This alone constitutes a significant numerical complication when the term is included. Furthermore, as we shall see in section 5.2, the term containing ν corresponds precisely to the friction kernel in the classical limit. For these reasons it is possible to describe the neglected term as a “quantum friction”.

Finally, while neglecting this term is a rather severe approximation, it is important to note that the statistical character of ν is identical regardless of whether one chooses to evolve from a partitioned initial condition, meaning that nothing is lost by this approximation when assessing the comparative impact of using the ESLE vs. the SLE. i.e. while both evolutions will be approximated, they will both be neglecting a term that is insensitive to the initial conditions.

4.3.1 Temperature Dependence

To model the LZ type sweep, we thermalise the system at some time $t_0 < 0$ in the past with the bias $\epsilon_0 = \kappa t_0$ and then evolve it in real time for $t > t_0$. For the purposes of achieving a quicker relaxation to the asymptotic state, the cut-off frequency $\omega_c = 25$ was chosen for all simulations. Unlike in the equilibrium case, where a large cut-off frequency was used to ensure the energy scale of the bath was always much greater than that of the spin, in the driven case in our simulations the system starts and ends with considerably stronger bias. As all the dynamical changes occur in a window around $t = 0$, the effect of reducing the cut-off frequency is to narrow the region where the state transitions may happen. In addition, we set $\Delta = 1$, which means that effectively all parameters of the system are scaled to units of Δ .

Fig. 4.4 shows an ESLE evolution of $\langle \sigma_z \rangle = \text{Tr}(\rho(t)\sigma_z) = \rho_{11}(t) - \rho_{22}(t)$ at finite temperature, where parameters were chosen such that the initial density matrix

approaches that of the LZ initial condition, $\rho(t_0) \approx \rho_0^{LZ}$ of Eq. (4.39), although the evolution still begins from a finite time in the past. We expect that from this initial condition the cross-time correlations (which rapidly attenuate with time) are suppressed when evolving from a regime where the bias field is initially much stronger than thermal effects. This limiting case therefore also serves as a check that the ESLE predicts evolutions consistent with partitioned methods.

One can see that at finite temperatures the asymptotic behaviour does not necessarily converge to the LZ result even at weak coupling. In addition, while the mean state of the system rapidly converges to its asymptotic limit, the amplitude of oscillations around this state, and their rate of decay appears dependent on temperature, with oscillation amplitude decaying slower at lower temperatures. We also observe that the mean value approaches the LZ value as the temperature is lowered. This can be explained by the final state (and its convergence to the LZ limit) being dependent on the size of the temporal region where the bias field is comparable to the strength of thermal fluctuations. This region, where $|\beta\kappa t| \lesssim 1$ is when thermal effects will have the greatest impact on the dynamics of the system, as elsewhere the bias field dominates the system evolution. Therefore, we should expect the asymptotic state at lower temperature to lie closer to the LZ limit. Unfortunately, the time required for oscillations to decay sufficiently to confirm this is much longer at lower temperature. Given the excessive computational cost of longer simulation times in the ESLE (see Section 4.4), in Fig. 4.4 the asymptotic states for the two lowest temperatures are extrapolated from (oscillating) data. From this we conclude that high temperatures (or slow sweeps) allow thermal effects to increase the asymptotic $\langle\sigma_z\rangle$ value away from $\langle\sigma_z\rangle_{LZ}$, consistent with earlier SSE results [83].

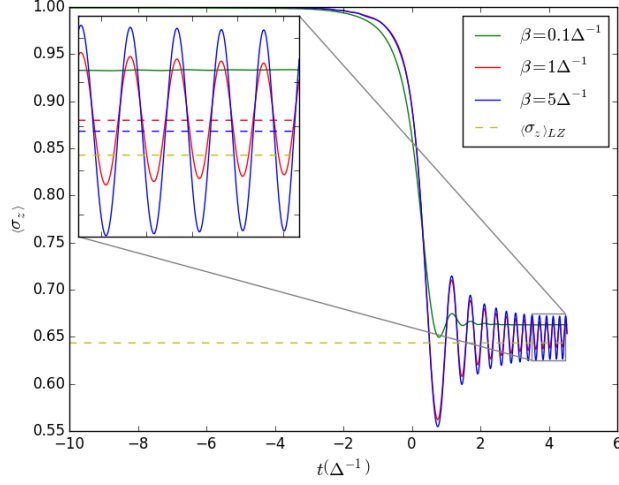


Figure 4.4: ESLE evolution of $\langle \sigma_z \rangle$ for a fast sweep with $t_0 = -6\Delta^{-1}$, $\kappa = 4\Delta^2$, and $\alpha = 0.05$, sampled over 3×10^5 runs, calculated at three values of the temperature. Red and blue dashed lines indicate extrapolated asymptotes for the two lowest temperature simulations. We observe that even at weak coupling the asymptotic value of $\langle \sigma_z \rangle$ deviates from the LZ expectation $\langle \sigma_z \rangle_{LZ}$, although lower temperature asymptotic states lie closer to the LZ limit.

4.3.2 Coupling-Strength Dependence

Fig. 4.5 (a) shows the real time dynamics of $\langle \sigma_z \rangle$ for the LZ sweep at different coupling strengths. Here simulations are started from a sufficiently small t_0 such that the calculated initial density matrices $\tilde{\rho}(t_0)$ are distinguishable from ρ_0^{LZ} . Comparing results in Fig. 4.5, we find that the bath coupling has two principal effects. First, oscillatory behaviour in the spin expectations is suppressed by increasing bath coupling, as expected. The asymptotic survival probability also increases, for the same reason as when increasing temperature. Indeed, stronger coupling allows thermal effects to have a stronger influence on the evolution lifting the $\langle \sigma_z \rangle$ asymptote. This phenomenon can also be understood as the system-bath coupling renormalising the characteristic frequency scale of oscillations, resulting in a quicker thermalisation. This scaling can be expressed in terms of the renormalised tunneling element [11, 176]:

$$\Delta_r = \Delta \left(\frac{\Delta}{\omega_c} \right)^{\frac{\alpha}{1-\alpha}} \quad (4.42)$$

Given this scaling is an argument based purely on renormalising the system Hamiltonian, we should expect it to hold regardless of the initial condition chosen. It should be noted here that the ESLE is at root a faithful representation for a particular kind of initial condition, and should not affect the dynamical properties of a system. Fig. 4.5 (b) shows the $\langle \sigma_z \rangle$ dynamics when time is scaled via this renormalised tunneling element, demonstrating that the curves of varying α scale on top of each other, hence serving as another consistency check that the ESLE produces physically reasonable results.

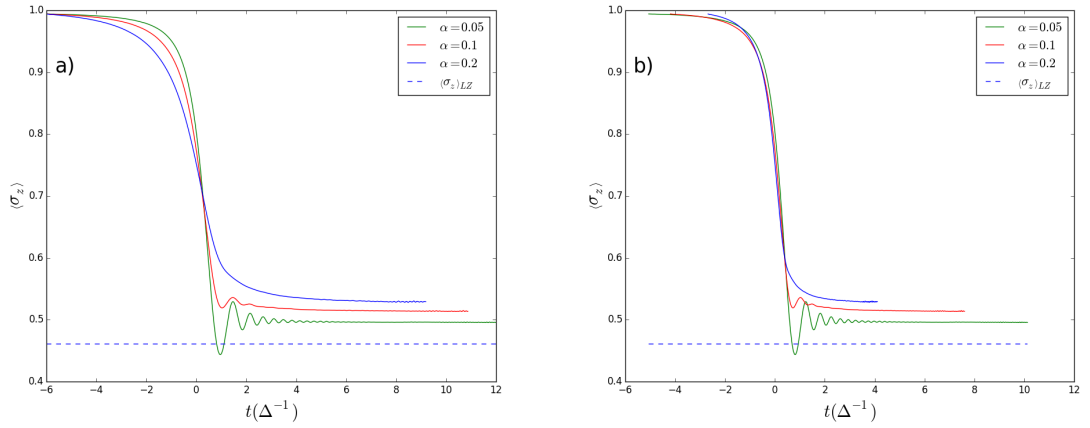


Figure 4.5: (a) Real time $\langle \sigma_z \rangle$ dynamics for the system with parameters $t_0 = -6\Delta^{-1}$, $\kappa = 5\Delta^2$, and $\beta = 0.2\Delta^{-1}$, after sampling with 5×10^5 runs. (b) The same dynamics are rescaled such that the curves of different α lie nearly on top of one another, demonstrating the spin-bath coupling renormalising the tunneling element of the two level system. Higher coupling strengths require more averaging to reach longer times, and hence have been truncated.

4.3.3 Comparison to Partitioned Evolution

We now compare the full ESLE to an SLE evolution purely in real time. Using the SLE, we may consider three different initial conditions:

- “SLE LZ”, where the system is evolved from the Landau-Zener initial condition:
 $\tilde{\rho}(t_0) = \rho_0^{LZ};$
- “SLE matched”, where the initial condition is that calculated from the averaged ESLE imaginary time evolution, i.e. from the exact reduced density matrix, $\tilde{\rho}(t_0) = \langle \bar{\rho}(\hbar\beta) \rangle_r = \frac{1}{Z_\beta} \text{Tr}_{\text{env}} [\exp(-\beta H_0)]$, obtained by solving Eq. (4.34); hence, this evolution differs from the exact ESLE only in that the cross-time correlation is set to zero;
- “SLE partitioned”, where the partitioned approximation is made to the initial state: $\tilde{\rho}(t_0) = Z^{-1} \exp[-\beta H_q(t_0)]$.

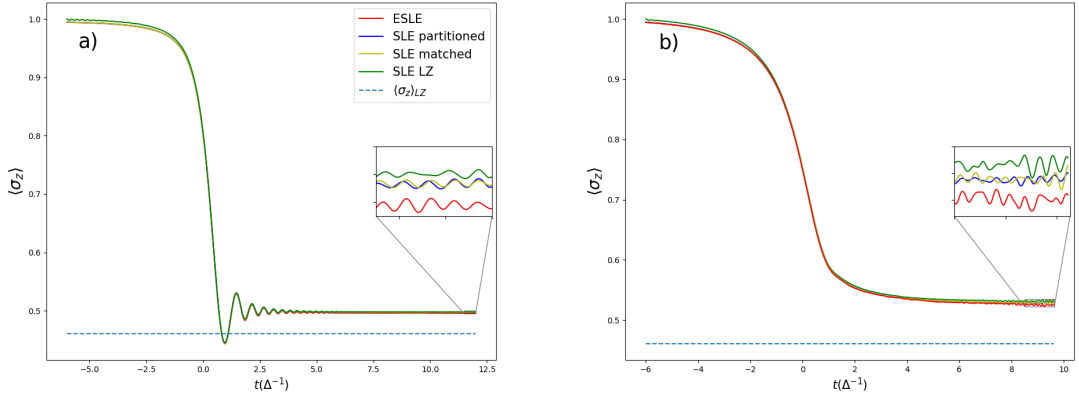


Figure 4.6: Real time spin dynamics for the system with parameters $t_0 = -6\Delta^{-1}$, $\kappa = 5\Delta$, $\beta = 0.2\Delta^{-1}$, sampled over 5×10^5 runs. (a) and (b) show $\langle \sigma_z \rangle$ at coupling strengths $\alpha = 0.05$, and $\alpha = 0.2$, respectively.

Fig. 4.6 shows the ESLE solution compared to these three cases of the SLE at both a) weak and b) strong coupling. There are several points to note here. The

first is that at these parameter settings the difference between the initial condition calculated by the ESLE and the naive initial condition used by the “SLE partitioned” approach is very small (but it is possible to make this difference in initial conditions more pronounced at higher temperatures and smaller initial biases). Otherwise, only the LZ initial condition is significantly different, as it exhibits initial oscillations (which are damped by stronger coupling), particularly in the coherence $\langle \sigma_x \rangle = \rho_{21}(t) + \rho_{12}(t)$. An example of this initial coherence oscillation at weak coupling is shown in Fig. 4.7, and an identical behaviour in decaying to zero is observed across all simulations.

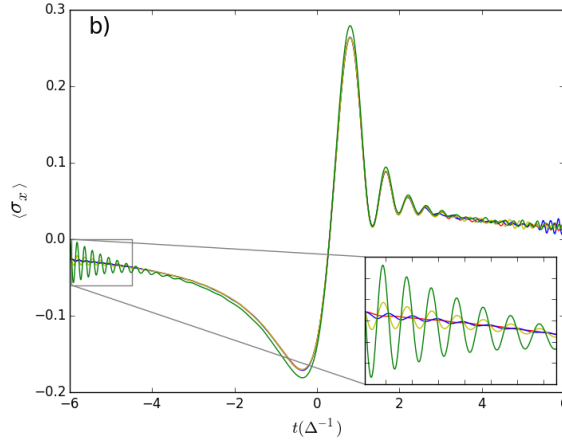


Figure 4.7: Real time spin dynamics of $\langle \sigma_x \rangle$ for the system with parameters $t_0 = -6\Delta^{-1}$, $\kappa = 5\Delta$, $\beta = 0.2\Delta^{-1}$, $\alpha = 0.05$ sampled over 5×10^5 runs. Except for the initial oscillations, the behaviour in all simulations appears identical.

We also see that the asymptotic states for all partitioned calculations are different to the ESLE at both coupling strengths. The partitioned simulations also display greater numerical instability, particularly at strong coupling, where a greater degree of averaging is required for confidence in the results.

At longer times we see a gap in $\langle \sigma_z \rangle$ develops between the ESLE and the three SLE simulations. To determine whether this gap is a real effect, or within the statistical error, the distributions of σ_z over individual runs were calculated at several time points

(see Fig. 4.8), and the error was estimated using the standard error:

$$\epsilon_\sigma = \frac{1}{\sqrt{N}} \sqrt{\sum_i (\sigma_z^i - \langle \sigma_z \rangle)^2} \quad (4.43)$$

where N is the number of runs and σ_z^i is the longitudinal spin from a single sample. Using this estimate, it was found that at long times there was a standard error of $\epsilon_\sigma = 5.9 \times 10^{-4}$ at $\alpha = 0.05$ and $\epsilon_\sigma = 1.2 \times 10^{-3}$ for $\alpha = 0.2$.

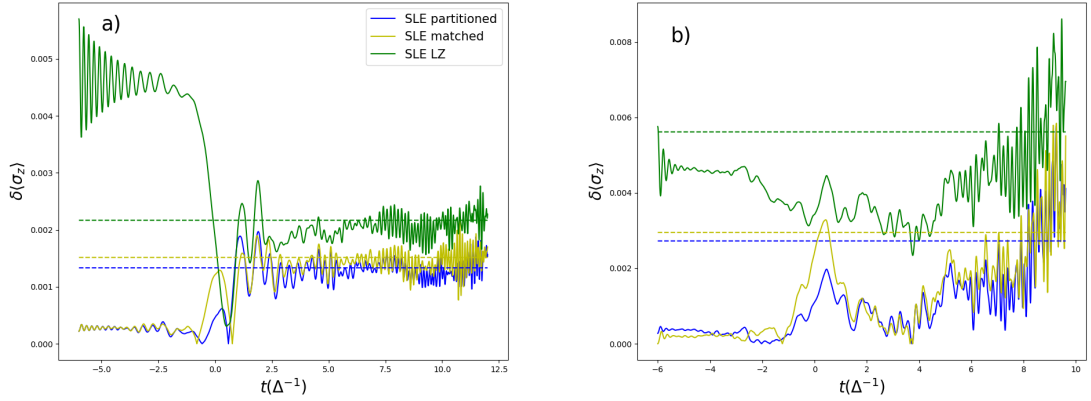


Figure 4.8: The evolution of the difference in $\langle \sigma_z \rangle$, calculated relative to the ESLE result of Fig. 4.6, at a) $\alpha = 0.05$, and b) $\alpha = 0.2$. The dashed lines indicate the mean value of the differences calculated using the final eighth of the simulation.

Comparison with the differences between the ESLE and other simulations (Fig. 4.8) clearly demonstrates that the gap is several times larger than the error at both coupling strengths. Conversely, the difference the partitioned and matched simulations almost overlay each other, with a smaller difference than the estimated error. This is surprising, as it suggests that even when the imaginary time evolution has little effect on the initial condition, the correlations it enforces impact the evolution at later times. The cross-time correlations are expected to have the largest effect at the start of the real-time dynamics due to the decay of the corresponding correlation kernels, but from the results we conclude this effect at short times accumulates to

produce the asymptotic gap.

Finally, Fig. 4.9 also serves to demonstrate the need for greater averaging to accurately model phenomena at longer times. The dynamics of the real system are reflected by a spreading of the distribution of single sample observables. This is how the mean and thus the physical density matrix evolve. A wider, flatter distribution will require a greater degree of sampling to reproduce the true mean value, hence longer evolutions requiring greater numbers of realisations.

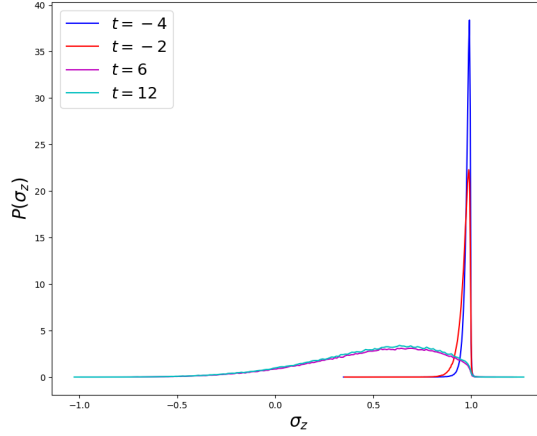


Figure 4.9: Distribution of σ_z at $t_0 = -6\Delta^{-1}$, $\kappa = 5\Delta^2$, $\beta = 0.2$ and $\alpha = 0.05$, sampled over 5×10^5 runs. As time progresses, the noise envelope drastically increases the width of the distribution, which requires more sampling to accurately calculate its mean.

4.4 Chapter Summary

In this chapter we have presented a numerical application of the exact ESLE to a driven spin-boson model. While there are no analytic predictions for evolutions from the exact initial density matrix presented, the numerical solution for the spin-boson system dynamics considered here using our exact partitionless method have been found to be extremely important as it can serve as a reference when comparing with

previous approximate calculations based on the partitioned approach. In the latter method, cross-correlations between system preparation (imaginary time evolution or thermalisation) and real time evolution are artificially missing. In this proof of concept for the method we have restricted ourselves to relatively short evolutions at high sweep speeds, to achieve quicker convergence of the results.

We have shown that for a simple system-bath coupling considered here only three Gaussian noises need to be generated: one for the imaginary time evolution that brings the entire system (the open system and the bath) to thermal equilibrium (initial preparation), and two noises for the real time evolution. The method presented here enabled us to generate these noises in such a way that all correlation functions are reproduced. We find, however, that small errors require a very large sampling set, i.e. up to and over 10^5 simulation runs are required to produce physically reasonable results.

As a sanity check of the method and its implementation, we first considered a real time evolution with a constant system Hamiltonian. One would expect that the real time evolution with the exact density matrix obtained after thermalisation in imaginary time should remain unchanged, and this was indeed found to be the case: we have seen that the ESLE predicts no change from initial conditions in its real time evolution. Partitioned SLE simulations from various initial conditions show relaxation, but on a timescale where we cannot reliably ascertain their steady state. Nevertheless, in the long time limit cross-time correlations die away, and both the ESLE and SLE will be evolved using noises with identical statistical properties, therefore we should expect the SLE simulation to converge to the ESLE result. At the same time, we find that great care is needed in achieving numerically acceptable results. This stems from the fact that the ESLE is a stochastic differential equation with multiplicative noise³, and is based on a first order finite difference approximation. This inevitably

³here we mean multiplicative simply in the sense that in the ESLE the noise is multiplied by ρ , in analogy to a multiplicative noise Langevin equation [177]

leads to a limit on the number of steps it may efficiently simulate before numerical instabilities dominate. As a possible future improvement of our method it may be prudent to implement a more sophisticated numerical scheme [178, 179]. This would help to converge the evolution with a larger timestep and hence allow access to longer timescales.

In addition to the equilibrium simulation, the non-equilibrium problem of a Landau-Zener sweep (in which the bias in the open system Hamiltonian is linearly driven) as an example of a fully non-equilibrium problem was also simulated. The exact ESLE simulation is compared specifically with the approximate (partitioned) SLE approach in which only real time evolution is considered from a chosen initial density matrix. We observe significant differences between partitioned evolutions and the ESLE, particularly at stronger coupling. We have found that the asymptotic behaviour of the ESLE in a Landau-Zener sweep protocol is *qualitatively* consistent with earlier results, showing that decreasing temperature and coupling strength brings the asymptotic solution for the survival probability closer to the known zero-temperature and zero-coupling result. At larger temperatures and coupling strengths the asymptotic state deviates significantly due to the presence of the bath, regardless of the initial condition used. In particular, even if one chooses to calculate the initial reduced density matrix exactly by thermalising the whole system, the cross-time correlations of the ESLE have a small (but observable) effect on the asymptotic state as compared to the SLE approach where this correlation is switched off.

These results highlight behaviours that may prove important in practical applications, particularly where quantum coherence is a resource, as there is a small (but persistent) difference between the ESLE and SLE predictions for driving away from equilibrium, particularly at short times. In addition, this approximation-free behaviour will affect the efficiency of quantum heat engines. It has already been shown that strong bath coupling diminishes their performance [63], and the results of ESLE

calculations confirm the well known fact that the state of the system when equilibrated with a heat reservoir is *not* the naive canonical state based on the isolated system Hamiltonian [180, 181].

Concluding, we stress that an important application of the ESLE is that it could serve as a test bed for verifying approximate analytical and numerical approaches. The essence of the ELSE is that only for a particular manifestation of the stochastic fields an analytical representation of the reduced density matrix (and hence a precise form of the corresponding effective Liouville equation describing a non-unitary evolution) is possible. Many such evolutions must be sampled in order to get the final physically meaningful result. Hence, the ELSE method is ultimately a numerical technique, but upon convergence it is capable of achieving an *exact* result. The fact that only numerical results are possible is not necessarily a disadvantage: even though an analytical solution with this method is out of reach, the fact that this method does provide an exact (albeit numerical) result is still important: firstly, one can obtain exact solutions for a particular Hamiltonian, and, secondly, the method can also be used to verify various approximate analytical theories.

Chapter 5

Classical Analysis

Gentlemen, there's lots of room left
in Hilbert space.

Saunders Mac Lane

In this chapter we shall explore some of the uses of the KvN formalism. Its principal advantage is the ability to import quantum mechanical techniques in the analysis of classical systems. In particular, in the second half of this chapter we shall demonstrate that a classical theory of self-adjoint operators may be used to determine the local entropic conservation properties of a probability distribution. This leads to a novel model in a restricted phase space where entropy is *oscillatory*. Some effort is then expended in the proper interpretation of this, concluding that the canonical distribution is one of the simplest functions with a well-defined, locally conserved entropy under Hamiltonian evolution.

More immediately, we derive a path integral formalism for generic classical systems based on the KvN propagator. This formulation allows for an analagous analysis of the Caldeira-Leggett model using the influence functional methodology employed in chapter 3. This proves to be an invaluable tool for establishing the correct classical limit of the ESLE. The naive approach previously used to take this limit obscures some

subtle detail, which can only be fully elucidated with reference to the KvN formalism.

5.1 The KvN path integral

As a preliminary to finding a classical equivalent to the ESLE, we must first derive the KvN path integral. This derivation follows the same procedure as in the quantum case, beginning with the KvN propagator

$$\hat{U}_{\text{cl}} = e^{-it\hat{K}} \quad (5.1)$$

where \hat{K} is given by Eq. (2.182). In the phase space representation this propagator is

$$U_{\text{cl}}(x_f, p_f, t_f; x_i, p_i, 0) = \left\langle x_f, p_f \left| e^{-it_f \hat{K}} \right| x_i, p_i \right\rangle. \quad (5.2)$$

We can perform a Trotter splitting, resulting in a similar expression to Eq. (2.19):

$$U_{\text{cl}}(x_f, p_f, t_f; x_i, p_i, 0) = \lim_{N \rightarrow \infty} \int dx_1 dp_1 \dots dx_{N-1} dp_{N-1} \prod_{j=0}^{N-1} \langle x_{j+1}, p_{j+1} | e^{-i\Delta \hat{K}} | x_j, p_j \rangle. \quad (5.3)$$

Considering a single term in this product, we have:

$$\langle x_{j+1}, p_{j+1} | e^{-i\Delta \hat{K}} | x_j, p_j \rangle = \langle x_{j+1}, p_{j+1} | \exp \left(-i \frac{\Delta}{m} \hat{\lambda} \hat{p} \right) \exp \left(-i \Delta \hat{\theta} V'(\hat{x}) \right) | x_j, p_j \rangle \quad (5.4)$$

which can be evaluated by inserting resolutions of unity

$$\exp \left(-i \Delta \hat{\theta} V'(\hat{x}) \right) | x_j, p_j \rangle = \int dx d\theta | x, \theta \rangle \langle x, \theta | x_j, p_j \rangle \exp \left(-i \Delta \theta V'(x) \right) \quad (5.5)$$

$$\langle x_{j+1}, p_{j+1} | \exp \left(-i \frac{\Delta}{m} \hat{\lambda} \hat{p} \right) = \int dp d\lambda \langle x_{j+1}, p_{j+1} | \lambda, p \rangle \exp \left(-i \frac{\Delta}{m} \lambda p \right) \langle \lambda, p |. \quad (5.6)$$

Using the overlaps specified by Eqs. (2.193) and (2.194), we obtain for Eqs. (5.5) and (5.6):

$$\exp\left(-i\Delta\hat{\theta}V'(\hat{x})\right)|x_j, p_j\rangle = \frac{1}{\sqrt{2\pi}} \int d\theta |x_j, \theta\rangle \exp(-i\Delta\theta V'(x_j) - i\theta p) \quad (5.7)$$

$$\langle x_{j+1}, p_{j+1} | \exp\left(-i\frac{\Delta}{m}\hat{\lambda}\hat{p}\right) = \frac{1}{\sqrt{2\pi}} \int d\lambda \exp\left(-i\frac{\Delta}{m}\lambda p_{j+1} + i\lambda x_{j+1}\right) \langle \lambda, p_{j+1} |. \quad (5.8)$$

Combining these together with Eq.(2.195) leads to the following expression for a single infinitesimal propagation

$$\langle x_{j+1}, p_{j+1} | e^{-i\Delta\hat{K}} | x_j, p_j \rangle = \frac{1}{(2\pi)^2} \int d\lambda_j d\theta_j \left[\exp\left(i\lambda_j \frac{\Delta}{m} \left(m \frac{x_{j+1} - x_j}{\Delta} - p_j\right)\right) \right. \quad (5.9)$$

$$\left. \exp\left(i\Delta\theta_j \left(\frac{p_{j+1} - p_j}{\Delta} + V'(x_j)\right)\right) \right]. \quad (5.10)$$

Note we have added a j subscript to the θ and λ variables in anticipation of inserting the appropriate resolutions of the identity. The overall propagator is therefore described by:

$$U_{\text{cl}}(x_f, p_f, t_f; x_i, p_i, 0) = \lim_{N \rightarrow \infty} \int \prod_{j=1}^{N-1} \left(\frac{dx_j}{\sqrt{2\pi}} \frac{dp_j}{\sqrt{2\pi}} \frac{d\lambda_j}{\sqrt{2\pi}} \frac{d\theta_j}{\sqrt{2\pi}} \right) \quad (5.11)$$

$$\times \exp\left(i \sum_{j=0}^{N-1} \lambda_j \frac{\Delta}{m} \left(m \frac{x_{j+1} - x_j}{\Delta} - p_j\right)\right) \exp\left(i \sum_{j=0}^{N-1} \Delta\theta_j \left(\frac{p_{j+1} - p_j}{\Delta} + V'(x_j)\right)\right). \quad (5.12)$$

In the limit we can once again describe this with a functional notation¹:

¹Note that we have cheated and moved directly to describing a time-dependent potential, which can be justified in the same way as in the quantum case

$$U_{\text{cl}}(x_f, p_f, t_f; x_i, p_i, 0) = \int_{x_i, p_i}^{x_f, p_f} \mathcal{D}x \mathcal{D}p \mathcal{D}\lambda \mathcal{D}\theta \, e^{iR} \quad (5.13)$$

$$R = \int_0^{t_f} dt \left[\lambda(t) \left(\dot{x}(t) - \frac{p(t)}{m} \right) + \theta(t) (\dot{p}(t) + V'(x, t)) \right] \quad (5.14)$$

The functional measure for each path variable is

$$\mathcal{D}f = \lim_{N \rightarrow \infty} \prod_n^N \frac{df_n}{\sqrt{2\pi}} \quad (5.15)$$

and compared to the quantum path integral, there is no factor of i causing the measure to fluctuate. For this reason, the KvN path integral is well behaved in the continuous limit.

The raw form of the KvN propagator is not particularly illuminating, but the integration over the ambiguity variables represents a product of delta functionals enforcing Hamilton's equations. We can see this most easily by returning to the discrete formulation. Specifically, consider the integration over λ_j :

$$\int d\lambda_j \exp \left(i\lambda_j \frac{\Delta}{m} \left(m \frac{x_{j+1} - x_j}{\Delta} - p_j \right) \right) = \frac{m}{\Delta} 2\pi \delta \left(m \frac{x_{j+1} - x_j}{\Delta} - p_j \right) \quad (5.16)$$

If this delta function is now integrated with respect to p_j , the propagator may be expressed with a reduced number of path variables. The functional measure is now

$$\mathcal{D}x \mathcal{D}\theta = \lim_{N \rightarrow \infty} \left(\frac{m}{2\pi\Delta} \right)^N \prod_n^N dx_n d\theta_n \quad (5.17)$$

while the propagator itself is:

$$U_{\text{cl}}(x_f, p_f, t_f; x_i, p_i, 0) = \int_{x_i, \dot{x}_i}^{x_f, \dot{x}_f} \mathcal{D}x \mathcal{D}\theta \exp \left[i \int_0^{t_f} dt \theta(t) (m\ddot{x}(t) + V'(x, t)) \right] \quad (5.18)$$

In this form, we see that the kernel of the path integral exponent enforces precisely the classical equations of motion; the KvN equivalent to the action in the quantum path integral. A useful feature of the KvN propagator is that it evolves both the classical wavefunction *and* the probability density (as they are governed by the same evolution equation). It is therefore possible to apply Eq.(5.18) to a localised probability density $\rho_0(x_i, p_i) = \delta(x_i - x_0) \delta(p_i - p_0)$:

$$\rho(x_f, p_f, t_f) = \int dx_i dp_i U_{\text{cl}}(x_f, p_f, t_f; x_i, p_i, 0) \rho_0(x_i, p_i) \quad (5.19)$$

$$= \int_{x_i, \dot{x}_i}^{x_f, \dot{x}_f} \mathcal{D}x \mathcal{D}\theta \exp \left[i \int_0^{t_f} dt \theta(t) (m\ddot{x}(t) + V'(t)) \right] \rho_0(x_i, p_i) \quad (5.20)$$

$$= \int dx_i dp_i \int_{x_i, \dot{x}_i}^{x_f, \dot{x}_f} \mathcal{D}x \rho_0(x_i, p_i) \delta[m\ddot{x}(t) + V'(t)] = \delta(x_f - x_{\text{cl}}(t_f)) \delta(\dot{x}_f - \dot{x}_{\text{cl}}(t_f)) \quad (5.21)$$

Hence, the particle remains localised with its trajectory $x_{\text{cl}}(t)$ described by the classical equation of motion. The KvN propagator in this special case constitutes what is undoubtedly the most excessive representation of single-particle Newtonian mechanics imaginable. Clearly, applying this formalism to single-particle classical mechanics returns nothing but the obvious, but by expressing the composite of an open system and its environment in this form, we are able to construct an influence functional to integrate out the environment explicitly.

Before continuing, there is a final point to address. Quantum mechanically, one can represent the thermal density matrix as an imaginary time path integral. In KvN mechanics, this really *is* a redundant exercise even for composite systems. This is because the classical Hamiltonian operator consists entirely of commuting operators, which kills the path integral. Formulating the classical density matrix explicitly, we have

$$\rho_\beta(x_N, p_N, x_0, p_0) = \left\langle x_N, p_N \left| e^{-\beta \hat{H}} \right| x_0, p_0 \right\rangle. \quad (5.22)$$

Trotter splitting the operator (using $\Delta = \beta/N$) produces

$$\rho_\beta(x_N, p_N, x_0, p_0) = \int dx_1 dp_1 \dots dx_{N-1} dp_{N-1} \prod_{n=0}^N \langle x_{n+1}, p_{n+1} | x_n, p_n \rangle e^{-\Delta H(x_n, p_n)} \quad (5.23)$$

$$\begin{aligned} &= \int dx_1 dp_1 \dots dx_{N-1} dp_{N-1} e^{-\Delta \sum_n H(x_n, p_n)} \prod_{n=1}^N \delta(x_{n+1} - x_n) \delta(p_{n+1} - p_n) \\ &= \delta(x_N - x_0) \delta(p_N - p_0) e^{-\beta H(q_0, p_0)} \end{aligned} \quad (5.24)$$

i.e. the classical density matrix is simply the canonical probability distribution, firmly demonstrating the futility of a path integral construction for a classical thermal density matrix. This may appear trivial, but it establishes an important difference between the classical and quantum treatments of the CL model. In the classical case, without an imaginary time path integral there is *no* possibility of introducing an imaginary time noise.

5.2 Classical Influence Functional

Using the path integral KvN formulation, it is possible to directly import many of the results derived for the quantum path integral. Principal among these is the ability to describe the reduced dynamics of an open system + environment amalgam with an equivalent influence functional formalism. For a global system described with canonical coordinates q, p and x, k , the total Hamiltonian is described by:

$$H_{\text{tot}}(q, x) = \frac{p^2}{2m} + \frac{k^2}{2m_k} + V_Q(q, t) + V_X(x, t) + V_{QX}(q, x, t) \quad (5.25)$$

initially described by the probability density

$$\rho_0^{\text{tot}} = \rho_Q(q_0, p_0) \rho_X(x_0, k_0) \quad (5.26)$$

using k as the momentum label for the X system. The classical reduced probability density is expressible as:

$$\begin{aligned} \rho_Q(q, p, t_f) &= \int \mathcal{D}q(t) \mathcal{D}\theta_Q(t) dq_0 dp_0 \mathcal{F}[q(t), p(t), \theta_Q(t)] \rho_Q(q_0; p_0) \\ &\times \exp \left(i \int_0^{t_f} dt \left[\theta_Q(t) \left(m \ddot{q}(t) + \frac{\partial V_Q}{\partial q}(q, t) \right) \right] \right) \end{aligned} \quad (5.27)$$

$$\begin{aligned} \mathcal{F}[q(t), p(t), \theta_Q(t)] &= \int dx_0 dk_0 dx_f dk_f \mathcal{D}x(t) \mathcal{D}\theta_X(t) \rho_X(x_0, k_0) \\ &\times \exp \left(i \int_0^{t_f} dt \left[\theta_X(t) \left(m_k \ddot{x}(t) + \frac{\partial V_{QX}}{\partial x}(q, x, t) + \frac{\partial V_X}{\partial x}(x, t) \right) + \theta_Q(t) \frac{\partial V_{QX}}{\partial q}(q, x, t) \right] \right) \end{aligned} \quad (5.28)$$

using $x(t_f) = x_f$ etc. The classical influence functional in abstract looks more formidable than its quantum equivalent, but this is largely due to a lack of convenient shorthand notation for the exponent integral. In practice this influence functional is much easier to evaluate, as we shall see when applying it to the Caldeira-Leggett model.

5.2.1 Classical Model

Since we are looking for the classical equivalent to the ESLE, we must evaluate the influence functional for the Caldeira-Leggett model. Our ultimate focus will be characterising the equivalent classical equation of motion for the reduced system, so we will forego many of the model generalisations made for the ESLE. The microscopic model in this case will involve a bath of internally decoupled, unit mass oscillators:

$$H_{\text{tot}} = H_Q(q) + \frac{1}{2} \sum_n (k_n^2 + \omega_n^2 x_n^2) - q \sum_n c_n x_n \quad (5.29)$$

We also implement the initial condition

$$\rho_0^{\text{tot}} = \rho_Q(q_0, p_0) \rho_\beta(\mathbf{x}_0, \mathbf{k}_0) \quad (5.30)$$

$$\rho_\beta(\mathbf{x}_0, \mathbf{k}_0) = \prod_n \frac{\beta \omega_n}{2\pi} \exp\left(-\frac{\beta}{2} (k_{0n}^2 + \omega_n^2 x_{0n}^2)\right) \quad (5.31)$$

i.e. the initial environment state is simply the bath of harmonic oscillators, neglecting the interaction contribution. It is actually possible to take the initial condition $\rho_0^{\text{tot}} = e^{-\beta H_{\text{tot}}}$ and include the interaction $-q \sum_n c_n x_n$ in ρ_β . In this case we would complete the square in the ρ_β exponent, redefining $x_{0n} \rightarrow x_{0n} - \frac{q_0 c_n}{\omega_n^2}$. This would result in an extra constant term $\beta \frac{q_0^2}{2} \sum_n \frac{c_n^2}{\omega_n^2}$ which could itself be cancelled by the inclusion of the counterterm found in Eq. (1.22). We have neglected this term, and any other term solely dependent on q , as the only effect due to these are modifications of the Q system potential and distribution, which are arbitrary to begin with. Critically, including this interaction, even when it is arbitrarily strong, does *not* introduce extra noise sources to the final result, or affect the correlations of these noises².

With this setup, we are able to insert the CL terms into Eq. (5.28), yielding:

$$\begin{aligned} \mathcal{F}[q(t), p(t), \theta_Q(t)] &= \prod_n \int dx_{0n} dk_{0n} dx_{nf} dk_{nf} \mathcal{D}x_n(t) \\ &\times \delta(\ddot{x}_n(t) + \omega_n^2 x_n(t) - c_n q(t)) \exp\left(i \int_0^{t_f} dt \theta_Q(t) c_n x_n(t)\right) \rho_\beta(x_{0n}, k_{0n}) \end{aligned} \quad (5.32)$$

where we have replaced the integrations over θ_X with their equivalent delta functionals. This delta functional will force the trajectory to obey $x_n(t) = x_n^{\text{cl}}(t)$, which solves the equation of motion:

$$\ddot{x}_n^{\text{cl}}(t) = -\omega_n^2 x_n^{\text{cl}}(t) + c_n q(t) \quad (5.33)$$

²It does however affect the translational invariance of the system Hamiltonian. While this is an important consideration, its consequences are not material to the arguments being made in this section.

The solution to this equation may be found in section 2.1.6, specifically Eq.(2.61). The classical equation of motion may therefore be taken as:

$$x_n^{\text{cl}}(t) = \frac{k_{0n}}{\omega_n} \sin(\omega_n t) + x_{0n} \cos(\omega_n t) + \frac{c_n}{\omega_n} \int_0^t dt' q(t') \sin(\omega_n(t - t')) \quad (5.34)$$

Inserting this into the influence functional,

$$\begin{aligned} \mathcal{F}[q(t), p(t), \theta_Q(t)] &= \prod_n \exp \left(i \int_0^{t_f} dt \theta_Q(t) \frac{c_n^2}{\omega_n} \int_0^t dt' q(t') \sin(\omega_n(t - t')) \right) \\ &\times \int dx_{0n} dk_{0n} \exp \left(i \int_0^{t_f} dt \theta_Q(t) c_n \left(\frac{k_{0n}}{\omega_n} \sin(\omega_n t) + x_{0n} \cos(\omega_n t) \right) \right) \rho_\beta(x_{0n}, k_{0n}) \end{aligned} \quad (5.35)$$

and using Eq. (5.31) to substitute for ρ_β , we find that the integrals over initial positions and momenta are of a Gaussian form. These integrations can be performed separately. The x_0 integration yields

$$\int dx_{0n} \exp \left(-\frac{\beta}{2} \omega_n^2 (x_{0n}^2 + 2Ax_{0n}) \right) = \sqrt{\frac{\beta}{2\pi\omega_n}} \exp \left(-\frac{\beta}{2} \omega_n^2 A^2 \right) \quad (5.36)$$

$$A = \frac{ic_n}{\beta\omega_n^2} \int_0^{t_f} dt \theta_Q(t) \cos(\omega_n t) \quad (5.37)$$

while the k_0 integration results in a similar term:

$$\int dk_{0n} \exp \left(-\frac{\beta}{2} (k_{0n}^2 + 2Bk_{0n}) \right) = \sqrt{\frac{\beta}{2\pi}} \exp \left(-\frac{\beta}{2} B^2 \right) \quad (5.38)$$

$$B = \frac{ic_n}{\beta\omega_n} \int_0^{t_f} dt \theta_Q(t) \sin(\omega_n t). \quad (5.39)$$

Combining these we obtain

$$\exp \left(-\frac{\beta}{2} (\omega_n^2 A^2 + B^2) \right) = \exp \left(-\frac{c_n^2}{2\omega_n} k_B T \int_0^{t_f} dt \int_0^t dt' \theta_Q(t) \gamma_n(t-t') \theta_Q(t') \right) \quad (5.40)$$

$$\gamma_n(t-t') = \frac{1}{\omega_n} (\cos(\omega_n t) \cos(\omega_n t') + \sin(\omega_n t) \sin(\omega_n t')) = \frac{1}{\omega_n} \cos(t-t'). \quad (5.41)$$

Collecting these results, we are able to express the influence functional as

$$\mathcal{F}[q(t), p(t), \theta_Q(t)] = e^{-\sum_n \frac{c_n^2}{2\omega_n} \Phi_n} \quad (5.42)$$

$$\begin{aligned} \Phi_n = & -2i \int_0^{t_f} dt \theta_Q(t) \int_0^t dt' q(t') \sin(\omega_n(t-t')) \\ & + k_B T \int_0^{t_f} dt \int_0^t dt' \theta_Q(t) \gamma_n(t-t') \theta_Q(t'). \end{aligned} \quad (5.43)$$

At this point we can take the continuum limit for the oscillators,

$$\sum_n \frac{c_n^2}{2\omega_n} f_n \rightarrow \int_0^\infty \frac{d\omega}{2\pi} I(\omega) \quad (5.44)$$

such that our final influence functional is given by:

$$\begin{aligned} \mathcal{F}[q(t), p(t), \theta_Q(t)] = & \exp \left[2i \int_0^{t_f} dt \theta_Q(t) \int_0^t dt' q(t') \frac{d\gamma(t-t')}{dt'} \right. \\ & \left. - \int_0^{t_f} dt \int_0^t dt' \theta_Q(t) k_B T \gamma(t-t') \theta_Q(t') \right] \end{aligned} \quad (5.45)$$

$$\gamma(t-t') = \int_0^\infty \frac{d\omega}{\omega\pi} I(\omega) \cos(t-t') \quad (5.46)$$

Once again our influence functional has a double integral in its exponent. At this juncture we will use the HS transformation, but we have a choice between transforming over only the square θ_Q term, or additionally incorporating the q variable. We will

consider the latter option in the next section, but for now we shall simply use

$$\exp \left(-\frac{1}{2} \int^{t_f} dt \int^{t_f} dt' \theta_Q(t) 2k_B T \gamma(t-t') \theta_Q(t') \right) = \left\langle \exp \left(-i \int_0^{t_f} dt \theta_Q(t) \eta_{\text{cl}}(t) \right) \right\rangle_r \quad (5.47)$$

$$\langle \eta_{\text{cl}}(t) \eta_{\text{cl}}(t') \rangle_r = 2k_B T \gamma(t-t'). \quad (5.48)$$

Putting all of this together, we find that the KvN propagator for a single trajectory is:

$$\tilde{U}_{\text{cl}} = \int_{q_0, \dot{q}_0}^{q_f, \dot{q}_f} \mathcal{D}q(t) \mathcal{D}\theta(t) \exp \left(i \int_0^{t_f} dt \theta(t) R(t) \right) \quad (5.49)$$

$$R(t) = m\ddot{q}(t) + V'(q, t) - \eta_{\text{cl}}(t) - 2 \int_0^t dt' q(t') \frac{d\gamma(t-t')}{dt'} \quad (5.50)$$

The final term in R can be integrated by parts:

$$\int_0^t dt' q(t') \frac{d\gamma(t-t')}{dt'} = q(t) \gamma(0) - q(0) \gamma(t) - \int_0^t dt' \dot{q}(t') \gamma(t-t') \quad (5.51)$$

the first two terms are pure functions of time of q , and hence can be absorbed into the arbitrary potential V , leaving only the friction term³. Substituting this back into the propagator and performing the path integral over $\theta(t)$ we obtain:

$$\tilde{U}_{\text{cl}} = \int_{q_0, \dot{q}_0}^{q_f, \dot{q}_f} \mathcal{D}q(t) \delta \left[m\ddot{q}(t) + V'(q, t) + 2 \int_0^t dt' \dot{q}(t') \gamma(t-t') - \eta_{\text{cl}}(t) \right] \quad (5.52)$$

This brings us to the ultimate result of this section, namely that the equation of motion for a single trajectory is a generalised Langevin equation:

$$m\ddot{q}(t) = -V'(q, t) - 2 \int_0^t dt' \dot{q}(t') \gamma(t-t') + \eta_{\text{cl}}(t) \quad (5.53)$$

³If we had included the interaction in our original thermal density, we would have had an extra term cancelling $q(0) \gamma(t)$ here, while including the counterterm in the open system Hamiltonian would cancel $q(t) \gamma(0)$.

In the particular case where $I(\omega) = D\omega$, we recover the archetypal Brownian motion:

$$\langle \eta_{\text{cl}}(t) \eta_{\text{cl}}(t') \rangle_r = 2k_B T D \delta(t - t') \quad (5.54)$$

$$\gamma(t) = D \delta(t) \quad (5.55)$$

5.2.2 Choice in the Hubbard-Stratonovich transformation

In order to derive the generalised Langevin equation, we specifically chose to perform the HS transform with respect to *only* the θ_Q variable. What if we had decided to implement the full HS transform? Returning to Eq.(5.45), we now apply the HS transform for both θ_Q and Q . The influence functional is then:

$$\mathcal{F}[q(t), p(t), \theta_Q(t)] = \left\langle \exp \left(i \int_0^{t_f} dt [-\theta_Q(t) \eta_{\text{cl}}(t) + q(t) \nu_{\text{cl}}(t)] \right) \right\rangle_r \quad (5.56)$$

$$\langle \eta_{\text{cl}}(t) \nu_{\text{cl}}(t') \rangle_r = -2i\Theta(t - t') \frac{d\gamma(t - t')}{dt} \quad (5.57)$$

$$\langle \nu_{\text{cl}}(t) \nu_{\text{cl}}(t') \rangle_r = 0. \quad (5.58)$$

The classical propagator for a single realisation is now expressible as:

$$\tilde{U}_{\text{cl}} = \int_{q_0, \dot{q}_0}^{q_f, \dot{q}_f} \mathcal{D}q(t) \delta[m\ddot{q}(t) + V'(q, t) - \eta_{\text{cl}}(t)] \exp \left(i \int_0^{t_f} dt q(t) \nu_{\text{cl}}(t) \right) \quad (5.59)$$

Just like in the heuristic classical limit of the ESLE, the equation of motion for an individual trajectory is a frictionless Langevin equation. The friction component has not vanished, but its influence on the expectations is to introduce a stochastic weighting on each trajectory. Clearly, the equations of motion for individual trajectories are affected by the presence or absence of a friction kernel, but the *expectations* of the two systems must be identical, provided the appropriate stochastic weighting is used in the averaging of the frictionless propagator. The classical limit in section 3.5.2 re-

produces the dynamics of a frictionless Langevin system, but obscures the non-trivial weighting on trajectories for expectations.

This interpretation is not entirely satisfying, as it implies a critical loss of information when taking the classical limit of the ESLE that must be restored with a post-hoc prescription for the weighting of trajectories. Clearly, it would be more desirable to formulate the ESLE in such a way that its classical equation of motion corresponds to Eq. (5.53) rather than Eq. (3.128). In the next section we shall detail precisely how to achieve this reformulation.

5.3 Alternative ESLE classical limit

Let us return to the quantum influence phase used to find the ESLE. In order to derive a classical limit consistent with a frictional Langevin equation, we must alter the form of the influence phase in Eq. (3.71) *before* employing the HS transform. In the case of the simplified CL model and initial condition provided by Eqs. (5.29) and (5.30), the influence phase reads:

$$\Phi[q, q'] = \frac{1}{2} \int_0^{t_f} dt \int_0^{t_f} dt' K^R(t - t') \epsilon(t) \epsilon(t') \quad (5.60)$$

$$+ 2i \int_0^{t_f} dt \int_0^{t_f} dt' [\theta(t - t') K^I(t - t')] \epsilon(t) y(t')$$

$$\epsilon(t) = q(t) - q'(t) \quad y(t) = \frac{1}{2} (q(t) + q'(t)) \quad (5.61)$$

$$K^R(t - t') = \int_0^\infty d\omega I(\omega) \coth\left(\frac{1}{2}\hbar\beta\omega\right) \cos(\omega t) \quad (5.62)$$

$$K^I(t) = - \int_0^\infty d\omega I(\omega) \sin(\omega t) \quad (5.63)$$

Rather than utilising the HS transformation for both ϵ and y , we perform it only over ϵ :

$$\Phi[q, q'] = \frac{1}{2} \int_0^{t_f} dt \int_0^t dt' \eta(t) \epsilon(t) + 2i \int_0^{t_f} dt \int_0^t dt' K^I(t-t') \epsilon(t) y(t') \quad (5.64)$$

where η has the usual autocorrelation $\langle \eta(t) \eta(t') \rangle_r = K^R(t-t')$. For the K^I term, we can integrate by parts with respect to t' . Defining $K^I(t) = \frac{d\gamma(t)}{dt}$, we obtain:

$$\begin{aligned} 2i \int_0^{t_f} dt \epsilon(t) \int_0^t dt' K^I(t-t') y(t') &= 2i \int_0^{t_f} dt \epsilon(t) [\gamma(t-t') y(t')]_0^t \\ &\quad - 2i \int_0^{t_f} dt \epsilon(t) \int_0^t dt' \gamma(t-t') \dot{y}(t') \dots \end{aligned} \quad (5.65)$$

The term $2i \int_0^{t_f} dt \epsilon(t) [\gamma(t-t') y(t')]_0^t$ when expressed in the original q, q' coordinates is decoupled, and just as in the classical case may be absorbed into the open system potentials for the forward and backward propagators separately. This allows us to make the substitution

$$2i \int_0^{t_f} dt \epsilon(t) \int_0^t dt' K^I(t-t') y(t') = -2i \int_0^{t_f} dt \epsilon(t) \int_0^t dt' \gamma(t-t') \dot{y}(t') \dots \quad (5.66)$$

As a result of these changes, the reduced density matrix for the system is evolved by an *effective* propagator, \tilde{U}_{eff} :

$$\tilde{\rho}_{t_f}(q; q') = \int d\bar{q} d\bar{q}' \tilde{U}_{\text{eff}}(q, q', t_f; \bar{q}, \bar{q}', 0) \tilde{\rho}_0(\bar{q}; \bar{q}') \quad (5.67)$$

$$\tilde{U}_{\text{eff}}(q, q', t_f; \bar{q}, \bar{q}', 0) = \int_{q(0)=\bar{q}}^{q(t_f)=q} \mathcal{D}q(t) \int_{q'(t_f)=q'}^{q'(0)=\bar{q}'} \mathcal{D}q'(t) \exp \left[\frac{i}{\hbar} S_{\text{eff}}[q(t), q'(t)] \right] \quad (5.68)$$

$$S_{\text{eff}}[q(t), q'(t)] = \int_0^{t_f} dt \left(L_Q[q(t)] - L_Q[q'(t)] + \eta(t) \epsilon(t) - 2\epsilon(t) \int_0^t dt' \gamma(t-t') \dot{y}(t') \right). \quad (5.69)$$

In this formulation, the propagator is no longer decoupled between the forward and backward trajectories⁴, preventing the straightforward identification of a classical limit as in Eq. (3.120). To address this, we express $L_Q[q(t)] - L_Q[q'(t)]$ in the sum-difference coordinates

$$S_{\text{eff}}[\epsilon(t), y(t)] = \int_0^{t_f} dt \left[m\dot{\epsilon}(t) \dot{y}(t) - V\left(y(t) + \frac{\epsilon(t)}{2}\right) + V\left(y(t) - \frac{\epsilon(t)}{2}\right) + \eta(t) \epsilon(t) - 2\epsilon(t) \int_0^t dt' \gamma(t-t') \dot{y}(t') \right]. \quad (5.70)$$

Note that this effective action is *exactly* of the form for the quantum-classical generator in Eq. (2.208) for $\kappa = 1$.

To obtain the classical result, we note that the average size of the fluctuating coordinate $\epsilon(t)$ will be proportional to \hbar . The crucial step in obtaining the classical limit is approximating \hbar as small before taking the limit:

$$V\left(y(t) + \frac{\epsilon(t)}{2}\right) - V\left(y(t) - \frac{\epsilon(t)}{2}\right) \approx V'(y(t)) \epsilon(t) \quad (5.71)$$

$$\eta \approx \eta_{\text{cl}} \quad (5.72)$$

which becomes exact in the $\hbar \rightarrow 0$ limit. Integration of the first term in the effective action by parts yields:

$$S_{\text{eff}}[\epsilon(t), y(t)] = \int_0^{t_f} dt \epsilon(t) \left[-m\ddot{y}(t) - V'(y(t)) + \eta_{\text{cl}}(t) - 2 \int_0^t dt' \gamma(t-t') \dot{y}(t') \right]. \quad (5.73)$$

⁴This propagator corresponds to an integro differential master equation for the density matrix in the coordinate basis. It was the introduction of the ν noise in the original derivation that enabled the decoupling of the forward and reverse propagators,

To perform the $\hbar \rightarrow 0$ limit, we must re-examine the path integral measure⁵ (e.g. Eq. (2.24) in its discrete form:

$$\mathcal{D}y(t) \mathcal{D}\epsilon(t) = \lim_{N \rightarrow \infty} \left(\frac{m}{2\pi\hbar\Delta} \right)^N \prod_n^N dy_n d\epsilon_n. \quad (5.74)$$

Making the substitution $\theta(t) = \frac{\epsilon(t)}{\hbar}$, the measure now reads:

$$\mathcal{D}y(t) \mathcal{D}\theta(t) = \lim_{N \rightarrow \infty} \left(\frac{m}{2\pi\Delta} \right)^N \prod_n^N dy_n d\theta_n \quad (5.75)$$

Comparison to Eq.(5.17) reveals this is the KvN measure. Furthermore, the effective propagator is now

$$\tilde{U}_{\text{eff}} = \int_{y_0, \dot{y}_0}^{y_f, \dot{y}_f} \mathcal{D}y(t) \mathcal{D}\theta(t) \exp \left(i \int_0^{t_f} dt \theta(t) R(t) \right) \quad (5.76)$$

$$= \int_{y_0, \dot{y}_0}^{y_f, \dot{y}_f} \mathcal{D}y(t) \delta[R(t)] \quad (5.77)$$

$$R(t) = m\ddot{y}(t) + V'(y, t) - \eta_{\text{cl}}(t) + 2 \int_0^t dt' \dot{y}(t') \gamma(t - t'). \quad (5.78)$$

There is now *no* \hbar dependence in this path integral⁶, and we have recovered the KvN propagator found in Eq.(5.52). This demonstrates that when the friction kernel is explicitly included in the quantum mechanical path integral, the classical limit corresponds exactly to the KvN path integral. The fact that the classical limit of the ESLE and the KvN path integral coincide is a valuable consistency check for both of these results.

⁵The Jacobian for the transformation of path variables to ϵ and y is unity.

⁶The effect of taking the $\hbar \rightarrow 0$ limit is $y(t) \rightarrow q(t)$, while the initial density matrix becomes the probability distribution $\tilde{\rho}_0(y; \theta) \rightarrow \tilde{\rho}_0(q)$

Aside: The Dynamical Origin of Irreversibility

Now we have firmly established the ESLE and its classical equivalent, it is worth taking a moment to consider its broader implications for the origins of irreversibility. I'll preface this section by saying that the arguments presented here are not novel, but the results we have obtained serve to emphasise them.

Reconciling irreversibility with Hamiltonian dynamics is not easy. The two descriptions have directly contradictory properties, but if physical laws are universal then they *must* be equivalent. Formally, irreversible systems may be described stochastically. Regardless of whether one chooses to consider individual trajectories or their expectation values, these descriptions break time reversal symmetry. In the quantum case this is expressed as a non-Hermitian Hamiltonian, while in the classical limit this corresponds to a velocity dependent friction kernel. In both cases these properties are irrevocably associated with the introduction of stochasticity. In order to establish that this description is consistent with Hamiltonian dynamics, a logically consistent route to a stochastic description is necessary.

The essential observation that it is possible to derive these properties from a Hamiltonian system is decades old [19], but incomplete. Starting from a Newtonian picture, one is forced to make some reasonable (but ultimately unjustified) assumptions about the initial conditions of the environment. This is very much an ad hoc motivation for introducing stochastic terms, but captures the essential idea that irreversibility is a consequence of the loss of information. As we have seen, the rigorous introduction of this concept (whether classically or quantum mechanically) requires a Hilbert space formalism. If one accepts this (and the attendant implication that physical laws are statistical in nature) then the divide between the description of open and closed systems (and their apparently contradictory properties) disappear.

A process appears irreversible precisely because we have marginalised out a part of

the system. Once we have discarded this information, our uncertainty as to the state of the system inevitably grows, *regardless* of which direction one chooses to evolve in time. It is impossible to spontaneously gain information- we can only gain it by putting (literal) work in⁷.

From this perspective the arrow of time is the arrow of ignorance- dynamics are time symmetric, but partial loss of information about a system results in an increase in uncertainty. We do not experience reality with perfect information, and this imposes the apparent temporal asymmetry we see in the world.

5.4 Classical Self-Adjoint Extensions and Entropy

Having gone to the trouble of expounding the Koopman von-Neumann theory, we can use it to perform further classical analysis. In particular, we can investigate entropy. Entropy is a critical descriptive component in physical theories, serving as a measure of irreversibility [182], the arrow of time [183–185], as well as the proper definition of equilibrium [186]. These examples emphasise that the physically meaningful content of entropy is contained in its time dynamics, rather than its absolute value. Given the vital role the dynamics of entropy plays in characterising physical systems, the conditions under which its conservation is mathematically guaranteed is of some interest.

Simply characterising entropy is however problematic, reflecting its anthropomorphic nature. It will not be found in any equation of motion, and the entropy one measures depends on the level of course graining in the phase space, and the variables one concerns themselves with [187]. Here we adopt the Gibbs measure of entropy for a probability density ρ

$$S = - \int dqdp \, \rho \ln(\rho) \tag{5.79}$$

⁷Landauer erasure is a canonical example of this.

which at equilibrium is equivalent to the thermodynamic entropy [188].

It is often stated that for a reversible process, the entropy change is zero. This is closely related to the notion that a Hamiltonian system is itself time reversible. The two ideas are often conflated into the notion that systems undergoing Liouvillian evolution conserve entropy. This is not in fact the case. Consider that if ρ is not explicitly time-dependent, it evolves according to the Liouville equation:

$$\dot{\rho} = \{H, \rho\} \quad (5.80)$$

Substituting the Poisson bracket directly into Eq. 5.79 and integrating by parts, one finds that the entropy production arises solely from boundary terms (e.g., for a system with box boundaries at q_{\pm}, p_{\pm}):

$$\begin{aligned} \dot{S} = & \left[\int dp \, \rho (\ln(\rho) + 1) \frac{\partial H}{\partial p} \right]_{q_-}^{q_+} - \left[\int dq \, \rho (\ln(\rho) + 1) \frac{\partial H}{\partial q} \right]_{p_-}^{p_+} \\ & + \left[\int dq \, \rho \frac{\partial^2 H}{\partial p \partial q} \right]_{p_-}^{p_+} - \left[\int dp \, \rho \frac{\partial^2 H}{\partial q \partial p} \right]_{q_-}^{q_+}. \end{aligned} \quad (5.81)$$

It is at this point that one generally assumes boundary conditions to kill these terms [189], but this is overly restrictive, and peculiar to both the dynamics and distribution one is evolving. This does not *in general* answer the question of entropy conservation. At the same time, it is difficult to assess how modifications, such as a restriction in phase space, would affect the entropy production of a system.

In order to answer the question of entropy conservation with the greatest possible generality, we shall employ the Koopman von-Neumann (KvN) formalism. In particular, we shall use this framework to equate entropy conservation to a classical self-adjoint evolution operator.

The theory of self-adjoint operators is closely bound to the evaluation of boundary conditions, and is vital in the resolution of apparent paradoxes in quantum mechanics

[190]. The theory also makes explicit connection to topological phases [191–193] and spontaneously broken symmetries [194–196]. More abstractly, self-adjointness assumes a vital role in novel approaches to proving the Riemann zeta hypothesis [197]. It is, in short, a useful piece of machinery, which may be applied to the problem of entropy conservation when alloyed to the KvN framework.

It may appear at first glance that this is a formal hammer cracking a classical nut, but formulating the problem in this way provides a key advantage; one may draw on the wealth of theory developed for operators on Hilbert space. In particular, the theory of self-adjoint operators allows one to make more general statements about entropy conservation. Explicitly, if \hat{K} is self-adjoint, then the Gibbs entropy will be conserved. In order to prove this however, we must first review the definition of a self-adjoint operator.

5.4.1 Self-Adjoint Operators

A self-adjoint operator \hat{A} has two important properties. First, it must be *Hermitian* [198]:

$$\langle \phi | \hat{A} \psi \rangle = \langle \hat{A} \phi | \psi \rangle \quad (5.82)$$

Meaning the action of the operator and its adjoint are identical $\hat{A} = \hat{A}^\dagger$. For bounded operators, the self-adjoint and Hermitian properties are identical. In the *unbounded* case (which is almost always true for physical observables) [198], an operator has a *domain* of states $\mathcal{D}(\hat{A})$ it is restricted to act upon.

A simple, illustrative example of this is to consider the quantum momentum operator $\hat{p} = -i\hbar \frac{d}{dq}$ on an interval of length L :

$$\langle \phi | \hat{p} \psi \rangle = -i\hbar \int_0^L dq \, \phi^* \frac{\partial \psi}{\partial q} \quad (5.83)$$

Integrating this term by parts we obtain

$$\langle \phi | \hat{p} \psi \rangle = -i\hbar [\phi^* \psi]_0^L + i\hbar \int_0^L dq \psi \frac{\partial \phi^*}{\partial q} \quad (5.84)$$

$$= \langle \hat{p} \phi | \psi \rangle - i\hbar [\phi^* \psi]_0^L. \quad (5.85)$$

Clearly, if \hat{p} is a Hermitian operator, the domain of states it acts on must be restricted only to those for which $[\phi^* \psi]_0^L = 0$. When

$$\mathcal{D}(\hat{p}) = \left\{ \psi, \frac{\partial \psi}{\partial q} \in L^2[0, L], \psi(0) = \psi(L) = 0 \right\} \quad (5.86)$$

we satisfy the Hermitian condition. In this sense, Hermiticity imposes a boundary condition on the states an operator may act on. Notice however that for the operator with the above domain, there is no restriction on ϕ^* . In other words

$$\mathcal{D}(\hat{p}^\dagger) = \left\{ \psi, \frac{\partial \psi}{\partial q} \in L^2[0, L] \right\}. \quad (5.87)$$

This is a problem, as it means the adjoint operator acts on a different space of states. In this case the spectral theorem breaks down, and there is no way to establish a unique one-to-one correspondence between an observable and an operator [22]. For this reason, Hilbert space theories require that an observable operator be *self-adjoint*. This is a stronger condition requiring that in addition to an operator and its adjoint having the same action, their domain is identical $\mathcal{D}(\hat{A}) = \mathcal{D}(\hat{A}^\dagger)$ [190, 194]. In this simple case we can remedy the deficiency by using the operator \hat{p}_λ , which has the same action but the domain

$$\mathcal{D}(\hat{p}_\lambda) = \left\{ \psi, \frac{\partial \psi}{\partial q} \in L^2[0, L], \psi(0) = e^{i\lambda} \psi(L) \right\}. \quad (5.88)$$

This domain satisfies the Hermitian condition when $\mathcal{D}(\hat{p}_\lambda) = \mathcal{D}(\hat{p}_\lambda^\dagger)$. The operator

\hat{p}_λ is therefore self-adjoint.

The self-adjoint condition is a vital component of any Hilbert space theory with the ambition of describing physics, and in the next section we shall demonstrate that a self-adjoint evolution operator conserves the Gibbs entropy.

5.4.2 Entropy Conservation Using The Self-Adjoint Property

Having defined self-adjointness, we now demonstrate that a self-adjoint Koopman operator guarantees entropy conservation. First, define the operator:

$$\hat{S}(t) = -\hat{\rho}_{\text{cl}}(t) \ln(\hat{\rho}_{\text{cl}}(t)) \quad (5.89)$$

$$\hat{\rho}_{\text{cl}}(t) = |\psi_{\text{cl}}(t)\rangle\langle\psi_{\text{cl}}(t)| \quad (5.90)$$

If \hat{K} is self adjoint, the evolution is unitary⁸ and

$$\text{Tr} [\hat{S}(t)] = \text{Tr} [e^{-i\hat{K}t} \hat{S}(0) e^{i\hat{K}^\dagger t}] = \text{Tr} [\hat{S}(0)]. \quad (5.91)$$

In the final equality, we have exploited the unitarity of time evolution (which implies \hat{K} is self-adjoint) and assumed the trace is finite. Having demonstrated the time independence of the trace \hat{S} , we now express it in the q, p basis:

$$\hat{S} = - \int dq dp \rho(q, p) |q, p\rangle\langle q, p| \ln \left(\int dq' dp' \rho(q', p') |q', p'\rangle\langle q', p'| \right) \quad (5.92)$$

where ρ is the probability density. Expanding the logarithm in a Taylor series yields:

$$\hat{S} = - \int dq dp \rho \ln(\rho) |q, p\rangle\langle q, p| \quad (5.93)$$

⁸ \hat{K} is not actually an observable, but as noted in section 2.3 for Stones' theorem to apply it must be self-adjoint.

$$\text{Tr} [\hat{S}] = - \int dq dp \rho \ln(\rho) = S \quad (5.94)$$

Thus, we establish that $\text{Tr} [\hat{S}]$ is the entropy, and is conserved for self-adjoint \hat{K} . Of course, the major question is *when* \hat{K} fulfils this condition. In the position basis we have

$$\hat{K} = -i \{H, \odot\}. \quad (5.95)$$

Using the definition of Hermiticity results in

$$\langle \phi | \hat{K} \psi \rangle = i \int dq dp \phi^* \left(\frac{\partial H}{\partial p} \frac{\partial \psi}{\partial q} - \frac{\partial H}{\partial q} \frac{\partial \psi}{\partial p} \right) \quad (5.96)$$

$$= i \left[\int dp \phi^* \psi \frac{\partial H}{\partial p} \right]_{q_-}^{q_+} - i \left[\int dq \phi^* \psi \frac{\partial H}{\partial q} \right]_{p_-}^{p_+} + \langle \hat{K} \phi | \psi \rangle \quad (5.97)$$

From which it follows that if \hat{K} is Hermitian, we require

$$\left[\int dp \phi^* \psi \frac{\partial H}{\partial p} \right]_{q_-}^{q_+} - \left[\int dq \phi^* \psi \frac{\partial H}{\partial q} \right]_{p_-}^{p_+} = 0. \quad (5.98)$$

\hat{K} is therefore self-adjoint for the domain of states which fulfil this condition *and* $\mathcal{D}(\hat{K}) = \mathcal{D}(\hat{K}^\dagger)$. This domain therefore constitutes all states for which entropy is conserved. Hence, the set of entropy conserving distributions for a given system can be ascertained by finding the conditions under which \hat{K} is self-adjoint.

5.4.3 von-Neumann Deficiency index theorem

One may still ask what the utility is in shifting the framing of the original problem to this formalism. The domain of a classical self-adjoint operator is equivalent to the full set of entropy-preserving boundary conditions, but the advantage of this perspective is that one may test whether an operator (which includes its domain of states) is self-

adjoint directly through a theorem of functional analysis, known as the *von Neumann deficiency index theorem* [22].

This theorem checks whether an operator is self-adjoint by considering the number of eigenfunctions in $\mathcal{D}(\hat{A}^\dagger)$ with imaginary eigenvalues, i.e. those $\psi \in \mathcal{D}(\hat{A}^\dagger)$ satisfying the equation:

$$\hat{A}^\dagger \psi_\pm = \pm i\eta \psi_\pm \quad (5.99)$$

where η is a positive, real-valued constant. The number of independent solutions for ψ_\pm are the *deficiency indices* n_\pm . These determine three possibilities for \hat{A}^\dagger [198, 199]:

$$\begin{aligned} n_+ = n_- = 0 & \quad \hat{A} \text{ is essentially self-adjoint} \\ n_+ = n_- \geq 1 & \quad \hat{A} \text{ has self-adjoint extensions} \\ n_+ \neq n_- & \quad \hat{A} \text{ has no self-adjoint extension} \end{aligned} \quad (5.100)$$

Here a self-adjoint extension is an operator \hat{A}_λ with the same action as \hat{A} , but whose domain has been modified such that $\mathcal{D}(\hat{A}_\lambda) = \mathcal{D}(\hat{A}^\dagger)$ to make it self-adjoint. Fig. 5.1 shows an example, demonstrating the domain modification made by the self adjoint extension.

The number of parameters required to characterise the self adjoint extensions is equal to n_\pm^2 [194]. This is exemplified by the quantum momentum operator on the interval $[0, L]$ with the domain given by Eq.(5.86). In this case $n_\pm = 1$ and the single parameter required to characterise the self adjoint extensions in this case is of course λ .

To summarise, using Koopman dynamics reduces the question of entropy conservation to checking if the Koopman operator \hat{K} is self-adjoint. Self-adjointness is tested using the deficiency index theorem, which allows one to find the complete set of states (represented by a classical wavefunction) in the domain of the Koopman operator's self-adjoint extensions. This corresponds to the most general boundary

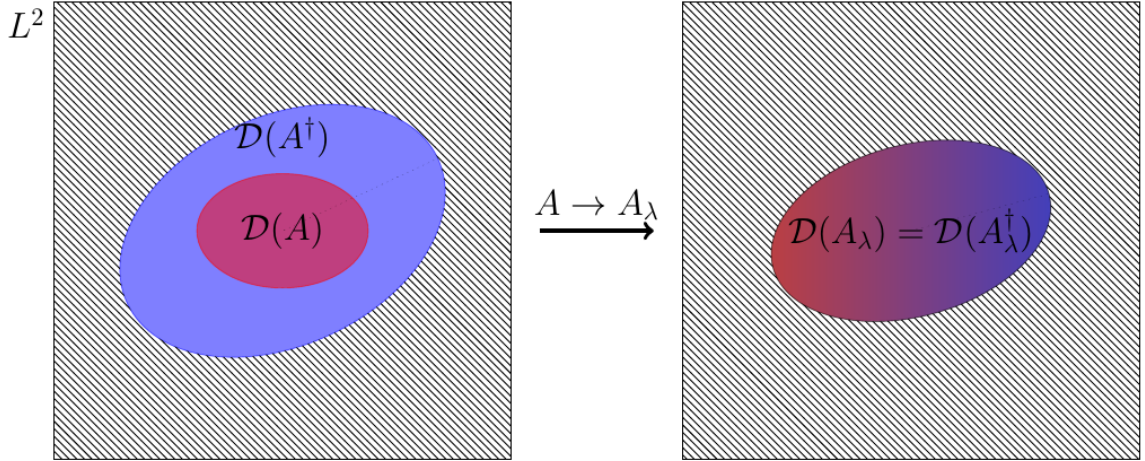


Figure 5.1: An operator A and its conjugate have different domains. A self adjoint extension modifies the domains of both operators such that they are identical.

conditions which conserve entropy for a given Hamiltonian. Establishing the self-adjoint extensions of an operator is itself a non-trivial exercise, but the analysis is most straightforward in a system where the Liouville operator is functionally dependent on only one phase space coordinate. Therefore in the following we will restrict ourselves to the specific example of a periodic system in action-angle coordinates, but emphasise that this is a choice of calculational convenience in applying more generic arguments.

5.4.4 An Example: Simple Periodic system

Consider a system whose coordinates can be canonically transformed into a space where one coordinate is cyclic, i.e. Hamiltonian that is functionally dependent on only coordinate. Any periodic system may be described by these canonical action-angle coordinates [1], but here we will take the simplest example of a harmonic oscillator:

$$H = \frac{1}{2} (q^2 + p^2) . \quad (5.101)$$

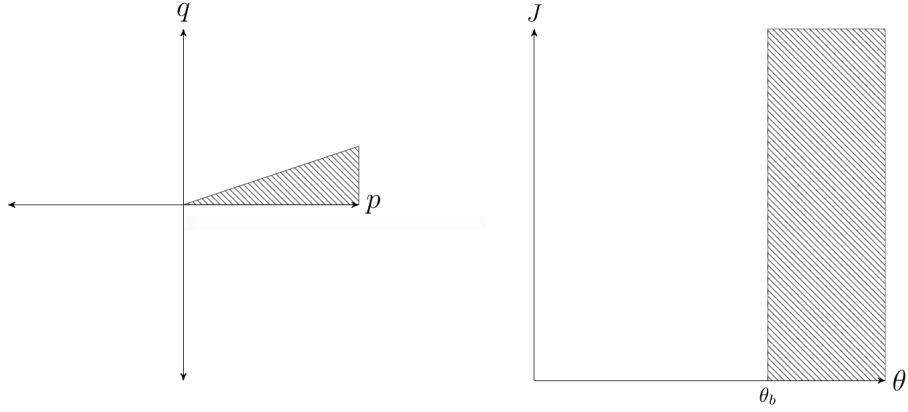


Figure 5.2: Example phase-space boundaries in (left) original coordinate system and (right) action-angle coordinates.

Using action-angle coordinates greatly simplifies the analysis, and can be achieved with the substitution:

$$q = \sqrt{2J} \sin(\theta) \quad p = \sqrt{2J} \cos(\theta). \quad (5.102)$$

We will stipulate that in the action-angle representation, the phase space has box boundaries, with $\theta \in [\theta_-, \theta_+]$ and $J \in [0, J_b]$ (see Figure 5.2). Given this is a canonical transform, the Poisson bracket structure for time evolution is retained in the action-angle variables. Since now $H = J$, the Koopman operator has a particularly simple representation in phase space

$$\hat{K} = -i \{H, \odot\} = -i \frac{\partial \odot}{\partial \theta}. \quad (5.103)$$

Applying the deficiency index theorem to the Koopman operator for the harmonic oscillator, we have:

$$\hat{K} \psi_{\pm} = \pm i \eta \psi_{\pm} \quad (5.104)$$

which after substitution with Eq. (5.103) reads

$$\frac{\partial \psi_{\pm}}{\partial \theta} = \mp \eta \psi_{\pm}. \quad (5.105)$$

One may solve this equation with a separation of variables to obtain:

$$\psi_{\pm}(\theta, J) = f_{\pm}(J) e^{\mp \eta \theta} \quad (5.106)$$

where $f_{\pm}(J)$ is an arbitrary function. Given this functional form, we check the number of solutions to ψ_{\pm} that lie in L^2 :

$$\begin{aligned} \int_{\theta_-}^{\theta_+} d\theta \int_0^{J_b} dJ |\psi_{\pm}|^2 &= \left(\int_0^{J_b} dJ f_{\pm}^2(J) \right) \left(\int_{\theta_-}^{\theta_+} d\theta e^{\mp 2\eta \theta} \right) \\ &= \mp \frac{1}{2\eta} \left(\int_0^{J_b} dJ f_{\pm}^2(J) \right) (e^{\mp 2\eta \theta_+} - e^{\mp 2\eta \theta_-}) \end{aligned} \quad (5.107)$$

Given f_{\pm} is arbitrary, any function satisfying $\int_0^{J_b} dJ f_{\pm}^2(J) < \infty$ forms part of a solution to ψ_{\pm} . From this we conclude that $n_+ = n_- = \infty$.

How should we interpret this result? For this system in action-angle coordinates, the Hermitian condition given by Eq. (5.98) is:

$$\int_0^{J_b} dJ [\phi^*(\theta, J, t) \psi(\theta, J, t)]_{\theta_-}^{\theta_+} = 0 \quad (5.108)$$

This can be satisfied by stipulating at each value of J the integrand is 0. In this case

$$[\phi^*(\theta, J, t) \psi(\theta, J, t)]_{\theta_-}^{\theta_+} = 0. \quad (5.109)$$

The most general way to enforce identical domains for \hat{K} and \hat{K}^\dagger is therefore

$$\forall J, t: \psi(\theta_+, J, t) = e^{i\beta(J)} \psi(\theta_-, J, t) \quad (5.110)$$

where $\beta(J)$ is an arbitrary real-valued function of J . For every value of J we have a one-parameter self-adjoint extension, and there are an infinite number of J values. This explains the result $n_+ = n_- = \infty$ and guarantees that in an unrestricted phase space, the probability distribution is continuous.

An interesting feature of this formulation is the admittance of classical wavefunctions acquiring an additional phase at each value of J as it circulates around phase space, illustrated in figure 5.3. One can ask if the choice of self-adjoint extension in this system is physically meaningful.

Assuming the system is in an eigenstate of the Koopman operator (where the eigenvalue spectrum may be J dependent):

$$\hat{K}\psi = \omega(J)\psi. \quad (5.111)$$

The most general solution to this equation is

$$\implies \psi(\theta, J, t) = e^{-i\omega(J)(\theta-t)}\chi(J) \quad (5.112)$$

where $\chi(J)$ is an arbitrary function in $L^2[0, J_b]$. Substituting this into Eq. (5.110) yields

$$\beta(J) = -\omega(J)(\theta_+ - \theta_-). \quad (5.113)$$

This demonstrates that the choice of self-adjoint extension determines the Koopman spectrum of the system. While this is not directly observable in expectations, it *is* gauge-invariant (in the sense of locally rotating a complete set of states). This is somewhat analogous to the Berry phase [200] (and its classical equivalent the Hannay angle [201]). Here however, the phase arises as an admissible, spontaneous phase change (for *each* value of J) after traversing the θ boundary, whose value depends on the choice of self-adjoint extension. In contrast, the geometric phase is due to

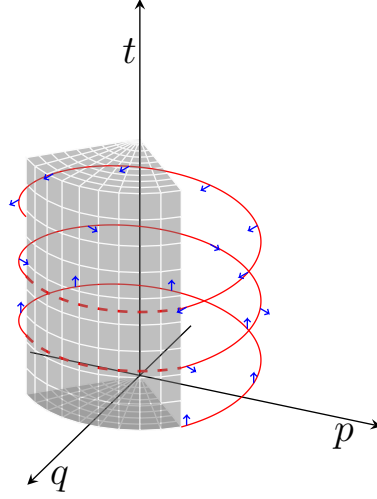


Figure 5.3: In the harmonic oscillator, time evolution is equivalent to rotation. Here the red curve represents the transport of $\psi(\theta_-, J, t)$ in time for a constant J . The blue arrows denote the phase due to the self-adjoint extension, where after each period an additional phase of $e^{i\beta(J)}$ is picked up.

adiabatic holonomic variation of the Hamiltonian parameters. It is well known that for the simple harmonic oscillator, the geometric phase change is zero [202], whereas in this case a phase may be acquired in the mismatch between the dynamical frequency (which is unity by construction) of the system, and the spectrum of the chosen self-adjoint extension.

Finally, we return to our original motivation of entropy conservation. Working from Eq.(5.81), we have:

$$\begin{aligned} \dot{S} &= - \int_0^{J_b} dJ \int_{\theta_-}^{\theta_+} d\theta \{H, \rho \ln(\rho)\} \\ &= \int_0^{J_b} dJ \int_{\theta_-}^{\theta_+} d\theta \frac{\partial(\rho \ln(\rho))}{\partial \theta} = \int_0^{J_b} dJ [\rho \ln(\rho)]_{\theta_-}^{\theta_+}. \end{aligned} \quad (5.114)$$

More generally, the time dependence of *any* observable in this simple system is given

by:

$$\langle \dot{f}(\theta, J) \rangle = \int_0^{J_b} dJ [\rho f(\theta, J)]_{\theta_-}^{\theta_+}. \quad (5.115)$$

Taking $\rho = |\psi|^2$, and applying the set of possible boundary conditions derived from the self adjoint extensions, one finds that as promised $\dot{S} = 0$.

In the quantum formalism, one finds that dissipative, irreversible phenomena are described by explicitly non-Hermitian Hamiltonians. This can either be instituted *a priori* as an effective model, or, as we have seen previously, derived as the equation of motion for a subsystem after part of the environment has been marginalised [137]. The fact that the requirement for entropy conservation is for the evolution operator to be self-adjoint opens up new possibilities for models which violate entropy and energy conservation. Specifically, we shall see in the next section that even for the harmonic oscillator, choosing a probability distribution that does not fulfil Eq. (5.110) violates entropy conservation.

5.4.5 Non-conserving Distribution In A Restricted Phase Space

Let us now consider what kind of boundary conditions lead to non self-adjoint Koopman operators, and non-conserved entropies. For the harmonic oscillator in action-angle coordinates, only θ plays a role in the dynamics. This logically should be the only coordinate where boundaries become physically relevant to the expectation of observables. This intuition is confirmed by the result in Eq. (5.110), demonstrating that only the θ coordinate is constrained at the boundaries. Additionally any system that can be cast in action angle coordinates must be periodic over the full 2π period of θ . i.e. if θ is unrestricted $\dot{S} = 0$ trivially.

We shall therefore take a probability distribution that is *not* globally θ symmetric, and consider its dynamics in a space where the range of θ is bounded to be *less* than 2π . There is of course a question over whether it is physical to impose a boundary

on momentum in this way, but let us proceed without concerning ourselves with this issue prematurely.

Consider the probability density (in the original q, p coordinates):

$$\rho(q, p) = Z^{-1} \exp(- (q^2 + qp + p^2)) \quad (5.116)$$

Expanding the exponent and expressing it in θ, J coordinates:

$$\rho(\theta, J) = Z^{-1} \exp(-2\gamma J - J\gamma \sin(2\theta)) \quad (5.117)$$

Note the multiplication by an arbitrary parameter γ in order to take advantage of differentiation by a parameter later. At the end of the calculation we'll set $\gamma = 1$. In this case it is easy to evaluate the microscopic entropy of the system:

$$\rho \ln(\rho) = Z^{-1} \exp(-\gamma J [2 + \sin(2\theta)]) (\ln(\mathcal{N}) - \gamma J [2 + \sin(2\theta)]) \quad (5.118)$$

$$= Z^{-1} \left(-\ln(Z) + \frac{\partial}{\partial \gamma} \right) \exp(-\gamma J [2 + \sin(2\theta)]) . \quad (5.119)$$

The entropy rate is therefore given by:

$$\dot{S} = Z^{-1} \left(-\ln(Z) + \frac{\partial}{\partial \gamma} \right) \int_0^{J_b} dJ \, e^{-2\gamma J} (\exp(-\gamma J \sin(2\theta_b)) - 1) \quad (5.120)$$

$$= Z^{-1} \left(-\ln(Z) + \frac{\partial}{\partial \gamma} \right) \left(\frac{e^{-2\gamma J_b}}{2\gamma} + \frac{1}{\gamma(2 + \sin(2\theta_b))} - \frac{1}{2\gamma} - \frac{e^{-\gamma J_b(2 + \sin(2\theta_b))}}{\gamma(2 + \sin(2\theta_b))} \right) \quad (5.121)$$

evaluating the derivative and setting $\gamma = 1$ yields:

$$\frac{\partial}{\partial \gamma} \left(\frac{e^{-2\gamma J_b}}{2\gamma} + \frac{1}{\gamma(2 + \sin(2\theta_b))} - \frac{1}{2\gamma} - \frac{e^{-\gamma J_b(2 + \sin(2\theta_b))}}{\gamma(2 + \sin(2\theta_b))} \right) \Big|_{\gamma=1} \quad (5.122)$$

$$= e^{-\gamma J_b(2 + \sin(2\theta_b))} \left(J_b + \frac{1}{2 + \sin(2\theta_b)} \right) - e^{-2\gamma J_b} \left(J_b + \frac{1}{2} \right) + \frac{1}{2} - \frac{1}{2 + \sin(2\theta_b)} \dots \quad (5.123)$$

The rate of entropy change is therefore

$$\dot{S} = -Z^{-1} (\ln(Z) + 1) \left[\frac{1}{2 + \sin(2\theta)} - e^{-J_b(2 + \sin(2\theta))} \left(J_b + \frac{1}{2 + \sin(2\theta)} \right) \right]_{\theta_-}^{\theta_+} \quad (5.124)$$

and for a general observable as a function of J :

$$f(J) = \sum_k c_k J^k \quad (5.125)$$

$$\langle f(\dot{J}) \rangle = Z^{-1} \sum_k c_k \left[\left(\frac{-1}{2 + \sin(2\theta)} \right)^{k+1} \frac{\partial^k}{\partial \gamma^k} \left| \frac{1 - e^{-\gamma J_b(2 + \sin(2\theta))}}{\gamma} \right|_{\theta_-}^{\theta_+} \right]. \quad (5.126)$$

It is clear from the functional forms of these expectations that non-conservation is due entirely to the θ boundary. Even when the phase space is unrestricted in J ($J_b \rightarrow \infty$ limit), there is a nonconserved term:

$$\lim_{J_b \rightarrow \infty} \dot{S} = -Z^{-1} (\ln(Z) + 1) \left(\frac{1}{2 + \sin(2\theta_+)} - \frac{1}{2 + \sin(2\theta_-)} \right). \quad (5.127)$$

Since time evolution is only determined by the θ coordinate, we can directly incorporate time into the θ argument

$$\frac{d}{dt} = -i\hat{K} = -\frac{\partial}{\partial \theta} \quad (5.128)$$

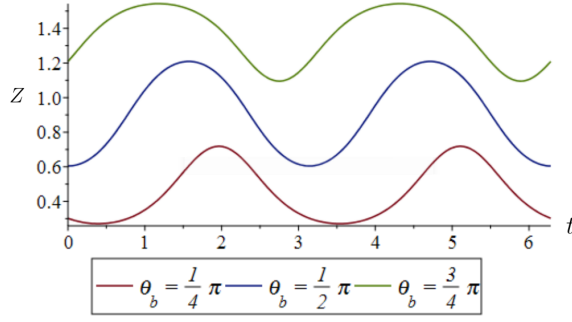


Figure 5.4: Normalisation time-dependence $\theta_+ - \theta_- = \theta_b$.

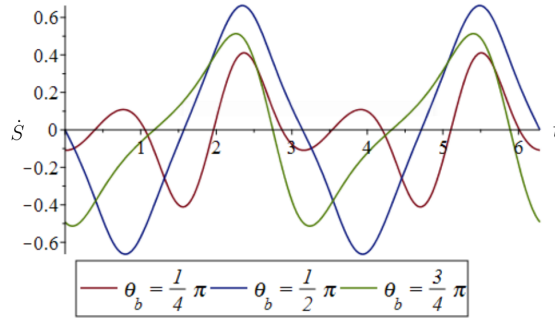


Figure 5.5: Entropy rate of change.

which implies that time and θ translation are identical, and hence

$$\rho(\theta, J, t) \equiv \rho(\theta - t, J). \quad (5.129)$$

This makes the normalisation of the distribution time-dependent:

$$Z(t) = \int_0^{J_b} dJ \int_{\theta_-}^{\theta_+} d\theta \rho(\theta - t, J) \quad (5.130)$$

$$= \int_0^{J_b} dJ \int_{\theta_- - t}^{\theta_+ - t} d\theta \rho(\theta, J). \quad (5.131)$$

As a result of restricting the phase space in the θ dimension, $Z(t)$, \dot{S} and $\langle \dot{J}(t) \rangle$ all acquire oscillatory dynamics, demonstrated in Figs. 5.4-5.6 (using $J_b \rightarrow \infty$).

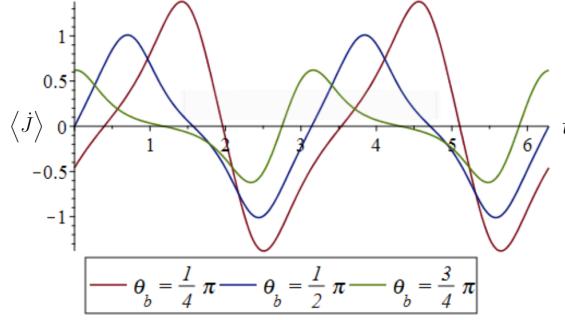


Figure 5.6: In this case we see $\langle J(t) \rangle$ is also a non-conserved quantity.

Note that the incorporation of time into the boundaries allows one to consider the interpretation that instead of physical dynamics on a restricted phase space, we have a static distribution with some information on an excluded region evolving dynamically.

Taking the distribution to be epistemological, at some point in time we check a region of phase-space and find no particle. Knowing the dynamics, the amount of information provided by that measurement about where the particle is *not* now depends on time. The probability distribution now possesses a rotating gap, as the difference in probability densities (and hence the non-conservation of entropy and J dependent expectations) for a given J depends on the boundary locations. This is illustrated in Fig. 5.7.

The non-conservation observed here demonstrates the importance of self-adjointness in a restricted phase space- while the evolution operator is apparently of Liouville form, the restriction in phase space has also limited the domain of classical states for which it preserves observable expectations. In this example, the failure of conservation is rather obvious in retrospect, but crucially the methods to find this distribution are more generally applicable to arbitrary Hamiltonians and phase-space topologies.

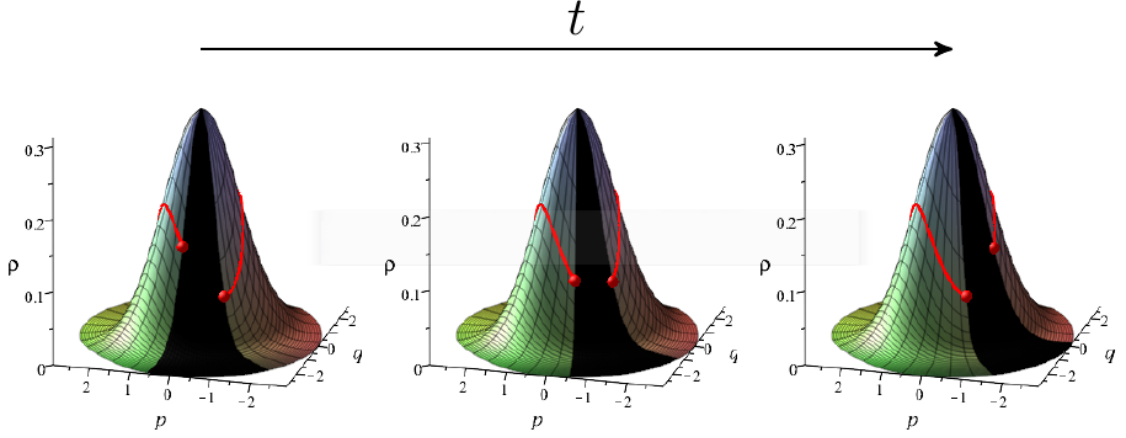


Figure 5.7: Distribution evolving with time. We can consider this as either a fixed excluded region with a rotating distribution, or a fixed distribution with an evolving excluded region. The red curve is a line of constant J , and the difference between the two boundary points on these curves is the essential contribution to entropy non-conservation.

5.5 Chapter Summary

The first part of the chapter derived the path integral formalism for the Koopman propagator, before applying the same methods used to derive the ESLE. This led to the identification of the classical Caldeira-Leggett model as a generalised Langevin equation. This analysis also served to properly establish the classical limit of the ESLE, and the origin of the missing structure when directly taking this limit. Ultimately, these derivations emphasised that stochastic, irreversible behaviour is a natural consequence of marginalising part of a composite system with reversible dynamics.

The latter half of the chapter employed the Koopman von-Neumann formalism to reduce the problem of entropy conservation in a classical system to identifying the self-adjoint extensions of the Koopman operator. In this way, one is able to explore the full range of admissible, physical probability distributions and phase space restrictions for a given system. Applying this technique to the harmonic oscillator, a relationship between the choice of self-adjoint extension, and the Koopman spectrum of the system

was determined. The self-adjoint extension manifests itself as a gauge-invariant phase.

This provides another example of the role self-adjoint extensions can play a role in subtle phenomena such as the determination of effective models of dissipation [203], as well as possessing more subtle links to phenomena such as topological phase transitions [204], and spontaneously broken symmetries [196]. These are typically quantum phenomena, but provided they are not explicitly dependent on a non-commutative algebra they should be reproducible classically. Specifically, there are already examples of classical Hamiltonians which emulate topological states [205]. Non-zero Chern numbers have been observed in models of biological structures [206], leading to the discovery of new classes of topological phonon bands [207]. In the ongoing exploration and analysis of these systems, both dissipative and topological, classical self-adjoint extensions should prove a useful tool.

In the case of a cyclic Hamiltonian, it was also found that any probability distribution continuous over the canonical angular boundary preserves entropy. By considering a restricted phase space, it was possible to formulate an effective model with the novel behaviour of oscillations in both entropy and energy. In this model, one finds that entropy production is no longer a measure of dynamical irreversibility, as the entropic oscillations suggest a local arrow of time which periodically reverses direction. This result was obtained with a simultaneous restriction of the q and p phase space coordinates, and one may legitimately claim this is an unphysical restriction. One would however obtain the same entropic oscillations with a *purely* spatial restriction⁹, and ultimately there are two ways to interpret the result.

The first approach is to consider the phase space restriction encoding some extra information about the system- i.e. a continuous measurement of that region (with a negative result). When we incorporate knowledge of the system probability distribution *and* its dynamics, the amount of information provided by this measurement will

⁹In fact, this analysis has been performed by a collaborator, but since it's not my work I haven't included it.

itself be dynamic, and naturally weight our expectations in a time dependent manner. As has been pointed out by Jaynes in his analysis of the Gibbs' paradox, entropy is an arbitrary measure, and its conservation is contingent on static information [187]. The Gibbs' paradox is *itself* only resolved by the uniform discarding of potentially accessible information (the distinguishability of particles), so it is not a surprise that phase space restrictions upset the usual statements on entropy.

Alternatively, one could consider that the self-adjoint extensions for a given region of phase space are those distributions enforcing *local* entropy conservation for that region. An interesting consequence of this is the interrogation of which distributions will conserve entropy within an *arbitrary* region of phase space.

For a cyclic system, entropy will be conserved regardless of the phase space boundaries when the distribution is a pure function $f(J)$. If we also impose the condition that $f(J) \ln(J)$ is well defined for all values of J , then one of the simplest non-trivial functions that meets these conditions is the canonical distribution $\frac{1}{Z} e^{-\beta H}$. While systems evolving under a cyclic Hamiltonian can always be considered dynamically equilibrated, the above consideration shows that thermodynamic equilibrium corresponds to a distribution which conserves the Gibbs entropy for an arbitrary region of phase space. In fact, it is relatively easy to show the thermal distribution will conserve local entropy for *any* Hamiltonian. It is in this sense that the Gibbs and Boltzmann entropies coincide at thermal equilibrium [208].

The example considered in this chapter has trivial dynamics, and therefore a simple set of entropy-preserving distributions, but the techniques used are more generally applicable. We emphasise that the entropic properties of a system are not determined purely via its dynamics, but may be engineered with a restricted phase space, rather than invoking non-Liouvillian, non-linear evolutions.

Chapter 6

Conclusions

Do I contradict myself? Very well
then I contradict myself; (I am
large, I contain multitudes.)

Walt Whitman

The initial goal of this body of work was a methodological extension of the influence functional formalism to account for a more general class of initial condition. The ESLE - detailed in chapter 3 - is the result of this. Its main advantage is that it projects out the environment *exactly*, without assuming that it is decoupled from the open system at the initial time. Moreover, at the initial time, prior to the action of a (possibly time dependent) perturbation, the ESLE method assumes the open system and the bath to be fully coupled and thermally equilibrated (although a broader class of initial conditions can also be introduced [64]). This means that the theory is able to faithfully capture the transient dynamics of driving the system away from the full system-environment equilibrium caused by any local (acting only on the open system) perturbation. The imaginary time part of the ESLE also provides a potential route to calculating partition functions for systems strongly coupled to their environment, as evidenced by the approximate calculation for the Hamiltonian of mean force (see

section 3.5.3).

Naturally, the ESLE is not without its shortcomings, the most obvious being that its domain of validity is restricted only to those systems well described by CL-type Hamiltonians. By describing only the dynamics of the reduced system, it also makes any information on the effect of the system on the environment inaccessible. One could for example drive an open system to an energy scale comparable to that of the reservoir. While the effect of this on the open system would be captured by the ESLE, it would be impossible to assess the state of the environment, which presumably would be severely affected by this driving.

Despite these problems, the arena of potential applications for the ESLE are rather broad. In the partitioned case, anharmonic bath models have been approximated by the CL Hamiltonian, leading to numerical schemes [209] for influence functional simulations of these systems [210]. There is no reason to suppose that the ESLE could not be extended in a similar manner. Properties such as thermal transport and entropy production through a spin system may also be calculated with the ESLE, allowing for the analysis of quantum heat engines. Analogous methods have already studied the heat exchange between an arbitrary system and a bath with Ohmic dissipation [74]. A particularly interesting development is the incorporation of a driven environment within the CL model [211]. Specifically, it is possible to take a Rubin model (consisting of two chains of oscillators coupled to a central system) [212] with a universal driving term and map this to the CL model. Examining the effects of this environmental driving on the stochastic properties of the ESLE would be an illuminating exercise.

Chapter 4 demonstrated the numerical feasibility of implementing the ESLE. In the two-state example studied, the effect of including imaginary time correlations was small, but demonstrably real! It is also not a forgone conclusion that just because the effect is small in a specific, simple system, that it is small in general. In the case of more complex systems, where the gap in energy scales between system and bath is

smaller, the true equilibrium distribution is potentially quite far from the partitioned approximation. The numerical implementation of the ESLE can also be improved, allowing more sophisticated applications of the method to analyse physical systems of interest. For example, the time evolution of more complex multi-state open systems interacting with bosonic fields (phonons and/or photons) [18]. This would require additional noises, necessitating a generalisation of the method to generate them. The addition of an effective non-Gaussian shot noise [167], or moving beyond the two-state system to a double-well potential [213, 214] also represent potential extensions, where phenomena such as metastable state stabilisation [215] become apparent. To achieve these aims, a more efficient algorithm for generating noises may also be necessary. In such a case, an extension of the spin-boson system into the incoherent regime and a study of its escape dynamics would be possible [216].

Beyond practical calculations, the ESLE embodies some more general principles. The inevitable breaking of time symmetry for a reduced system has been discussed previously, but the model also touches on the necessity of memory in thermalisation (a point raised in section 1.3). The relevance of this profound result to the ESLE is clear, given that it is explicitly constructed from an initially thermal state, and the noises one obtains are intrinsically non-Markovian. The key point is that deriving the noise correlations directly from a microscopic model guaranteed non-Markovianity. This important feature is missed in descriptions with *a priori* stochastic terms, and reflects the fact that there is no thermalisation without correlations!

The physical insight provided by the ESLE and similar equations are a consequence of the power of the influence functional formalism. This was in large part the motivation for chapter 5. By importing influence functionals into a KvN description, it was possible to derive a generalised Langevin equation directly from the CL model. This classical limit also reveals some insight into the categorisation of noises, and their effect on system dynamics. The ESLE is a stochastic differential equation, with

noise that is necessarily always multiplicative. The presence of this type of noise in classical systems (that is, a Langevin equation with multiplicative noise) gives rise to absorbing states and to noise induced phase transitions [177]. On this basis, one might expect to observe analagous behaviours in the ESLE, but this conflates the Langevin and Liouville equation of motion. By taking the classical limit, we find that the multiplicative noise in a Liouville equation corresponds to a Langevin equation with additive noise¹.

Incorporating quantum techniques into classical analysis is not limited to influence functionals, and can be used to develop new methodologies. The use of classical self-adjoint extensions and their relation to entropy conserving distributions is an example of this. This approach has resulted in a model with novel entropic properties, but its physical relevance is unclear. At this moment in time explorations of KvN theory are scarce, but it is clear that there remains a great deal to discover. My own interests here are oriented towards the quantisation of the theory, and quantum-classical transitions. In particular, the issue of distinguishability may be illuminated with the use of KvN dynamics. Classical particles are distinguishable, quantum particles are not. It seems to me that while the effect of this property on statistics is well established, distinguishability itself is poorly characterised. Nevertheless, this property must originate in the commutative structure of each theory, and a comparative analysis between the two may help to explicate it.

What ultimate conclusions can be drawn from the work presented here? Inevitably, this thesis is ludicrously specific, but I would hazard (more in hope than expectation) that it brushes against some more profound concepts. Chief among these is the necessity of a statistical interpretation of *all* physics. This idea was famously articulated in Max Born’s Nobel speech: “Ordinary mechanics must also be statistically formulated: the determinism of classical physics turns out to be an illusion, it is an idol, not an

¹This actually depends on the precise functional form of the coupling, but when $\hat{f} = \hat{q}$, this is true.

ideal in scientific research” [217].

Randomness is not an *ad hoc* model addition, but an essential, irreducible component in our description of reality. Its existence always reflects imperfect information, whether that is due to unobserved interactions with other systems, or a fundamentally non-commutative algebraic structure. My own motivation for this opinion² is the unified picture of physics provided by the Hilbert space formalism, combined with the Hubbard-Stratonovich transformation’s rigorous introduction of stochasticity.

Open quantum systems are such a fascinating area of physics partly because they contain multitudes - disparate phenomena that must be accounted for in a holistic way. This is only enriched by the realisation that classical systems can be described in the same manner. Dissipative quantum and classical phenomena sometimes appear to be distant cousins, related solely by analogy and inspiration. It is only when we combine their representations that it becomes possible to see that the mechanisms underlying both are fundamentally identical.

More generally, I think it is remarkable that we live in a world that is predictable. It appears that all an element of reality can do is exist and move, yet this is enough. All the variety and complexity on display in nature follows from stuff existing, and a set of rules about how it should move. In my mind, it seems almost perverse that more abstract qualities are not required. Furthermore, these rules are not only comprehensible, but compactly codifiable! Describing the observed world with linear models is not only unreasonably effective [218], but absurdly rich [219]. There is still much in open systems (let alone nature) that eludes explanation, but we should count ourselves fortunate. We know *something*, which as any infinitesimal analyst will tell you, is more than nothing.

²and at the end of the day it is just that, an opinion.

References

- [1] Herbert Goldstein. *Classical Mechanics*. Pearson, Essex, 3 edition, 2014.
- [2] V. I. Arnold. *Mathematical Methods of Classical Mechanics*. Springer New York, New York, NY, 1989.
- [3] John Dryden. *Mac Flecknoe (The Merrill literary casebook series)*. Merrill, 1970.
- [4] U. Weiss. *Quantum dissipative systems*. World Scientific, Singapore, 2009.
- [5] P. Hänggi, P. Talkner, and M. Borkovec. Reaction-rate theory: fifty years after Kramers. *Rev. Mod. Phys.*, 62:251, 1990.
- [6] V.I. Mel’nikov. The Kramers problem: Fifty years of development. *Physics Reports*, 209(1-2):1–71, 1991.
- [7] K.W.H. Stevens. The wave mechanical damped harmonic oscillator. *Proc. Phys. Soc.*, 72(6):1027, 1958.
- [8] E. Kerner. Note on the forced and damped oscillator in quantum mechanics. *Canadian J. Phys.*, 36(3):371–377, 1958.
- [9] E. Kanai. On the quantization of the dissipative systems. *Prog. Th. Phys.*, 3(4):440–442, 1948.

- [10] I.R. Senitzky and A Rutherford. Dissipation in Quantum Mechanics. The Harmonic Oscillator. *Phys. Rev.*, 119(2):670–679, 1960.
- [11] A.J. Leggett, S. Chakravarty, A.T. Dorsey, Matthew Fisher, Anupam Garg, and W. Zwerger. Dynamics of the dissipative two-state system. *Reviews of Modern Physics*, 59(1), 1987.
- [12] R. Schmidt, M. F. Carusela, J. P. Pekola, S. Suomela, and J. Ankerhold. Work and heat for two-level systems in dissipative environments: Strong driving and non-markovian dynamics. *Phys. Rev. B*, 91:224303, Jun 2015.
- [13] Rui Sampaio, Samu Suomela, Rebecca Schmidt, and Tapio Ala-Nissila. Quantifying non-markovianity due to driving and a finite-size environment in an open quantum system. *Phys. Rev. A*, 95:022120, Feb 2017.
- [14] S. Camalet and R. Chitra. Enhanced decoherence in the vicinity of a phase transition. *Phys. Rev. Lett.*, 99:267202, Dec 2007.
- [15] Juliana Restrepo, R. Chitra, S. Camalet, and Émilie Dupont. Effect of a gap on the decoherence of a qubit. *Phys. Rev. B*, 84:245109, Dec 2011.
- [16] R. J. Schoelkopf and S. M. Girvin. Wiring up quantum systems. *Nature*, 451(7179):664–669, 2008.
- [17] Denys I. Bondar, Wing-Ki Liu, and Misha Yu. Ivanov. Enhancement and suppression of tunneling by controlling symmetries of a potential barrier. *Phys. Rev. A*, 82:052112, Nov 2010.
- [18] Breuer Heinz-Peter and Francesco Petruccione. *The Theory of Open Quantum Systems*. OUP, Oxford, 2007.
- [19] J.S. Zwanzig. *Nonequilibrium Statistical Mechanics*. OUP, Oxford, 2001.

- [20] Ruixue Xu and YiJing Yan. Theory of open quantum systems. *The Journal of Chemical Physics*, 116(21):9196–9206, 2002.
- [21] Matthew Tyler. Absolute promotion and the condition on clitic hosts in choctaw. *Natural Language & Linguistic Theory*, 2018.
- [22] John von Neumann. *Mathematical Foundations of Quantum Mechanics*. Princeton University press, Princeton, 1932.
- [23] Karl Kraus, A. Böhm, J. D. Dollard, and W. H. Wootters, editors. *States, Effects, and Operations Fundamental Notions of Quantum Theory*. Springer Berlin Heidelberg, 1983.
- [24] Herbert B. Callen and Theodore A. Welton. Irreversibility and generalized noise. *Phys. Rev.*, 83(1):34–40, 1951.
- [25] Melvin Lax. Formal Theory of Quantum Fluctuation From a Driven State. *Phys. Rev.*, 129(5), 1963.
- [26] G. W. Ford, M. Kac, and P. Mazur. Statistical Mechanics of Assemblies of Coupled Oscillators. *J. Math. Phys.*, 6(4):504–515, 1965.
- [27] Rafael Benguria and Mark Kac. Quantum Langevin Equation. *Phys. Rev. Lett.*, 46(1-4), 1981.
- [28] Albert Schmid. On a quasiclassical Langevin equation. *Journal of Low Temperature Physics*, 49(5-6):609–626, 1982.
- [29] Katja Lindenberg and Bruce West. Statistical properties of quantum systems: The linear oscillator. *Phys. Rev. A*, 30(1):568–582, 1984.

- [30] Emilio Cortes, Bruce West, and Katja Lindenberg. On the generalized Langevin equation: Classical and quantum mechanical. *J. Chem. Phys.*, 82(6):2708–2717, 1985.
- [31] G. Lindblad. On the generators of quantum dynamical semigroups. *Communications in Mathematical Physics*, 48(2):119–130, jun 1976.
- [32] R. Alicki and K. Lendi. *Quantum Dynamical Semigroups and Applications*. Springer, 2007.
- [33] H M Wiseman. Quantum trajectories and quantum measurement theory. *Quantum and Semiclassical Optics: Journal of the European Optical Society Part B*, 8(1):205, 1996.
- [34] J. Echanobe, A. del Campo, and J. G. Muga. Disclosing hidden information in the quantum zeno effect: Pulsed measurement of the quantum time of arrival. *Phys. Rev. A*, 77:032112, Mar 2008.
- [35] R Alicki. The quantum open system as a model of the heat engine. *Journal of Physics A: Mathematical and General*, 12(5):L103, 1979.
- [36] Karl Blum. *Density matrix theory and applications*. Plenum Press, New York, 1996.
- [37] A. G. Redfield. On the theory of relaxation processes. *IBM J. Res. Dev.*, 1(1):19–31, January 1957.
- [38] Chris Fleming, N I Cummings, Charis Anastopoulos, and B L Hu. The rotating-wave approximation: consistency and applicability from an open quantum system analysis. *Journal of Physics A: Mathematical and Theoretical*, 43(40):405304, sep 2010.

- [39] W. Thomas Pollard and Richard A. Friesner. Solution of the redfield equation for the dissipative quantum dynamics of multilevel systems. *The Journal of Chemical Physics*, 100(7):5054–5065, 1994.
- [40] Hyun Keun Lee, Sourabh Lahiri, and Hyunggyu Park. Nonequilibrium steady states in langevin thermal systems. *Phys. Rev. E*, 96:022134, Aug 2017.
- [41] Ralf Metzler, Eli Barkai, and Joseph Klafter. Anomalous diffusion and relaxation close to thermal equilibrium: A fractional fokker-planck equation approach. *Phys. Rev. Lett.*, 82:3563–3567, May 1999.
- [42] Julian Lee. Derivation of markov processes that violate detailed balance. *Phys. Rev. E*, 97:032110, Mar 2018.
- [43] Goran Lindblad. Brownian motion of a quantum harmonic oscillator. *Reports on Mathematical Physics*, 10(3):393 – 406, 1976.
- [44] H. M. Wiseman and W. J. Munro. Comment on “dissipative quantum dynamics with a lindblad functional”. *Phys. Rev. Lett.*, 80:5702–5702, Jun 1998.
- [45] Bassano Vacchini. Quantum optical versus quantum brownian motion master equation in terms of covariance and equilibrium properties. *Journal of Mathematical Physics*, 43(11):5446–5458, 2002.
- [46] Dmitry V. Zhdanov, Denys I. Bondar, and Tamar Seideman. No Thermalization without Correlations. *Physical Review Letters*, 119(17):1–6, 2017.
- [47] A.O. Caldeira and A.J. Leggett. Path integral approach to quantum Brownian motion. *Physica A*, 121(3):587–616, 1983.
- [48] M. Rosenau da Costa, A. O. Caldeira, S. M. Dutra, and H. Westfahl. Exact diagonalization of two quantum models for the damped harmonic oscillator. *Phys. Rev. A*, 61:022107, Jan 2000.

- [49] R.P. Feynman and A.R. Hibbs. *Quantum Mechanics and Path Integrals*. Dover, New York, 2010.
- [50] J.S. Schulman. *Techniques and Applications of Path Integration*. Dover, New York, 1981.
- [51] P. Hänggi. Dissipative tunneling. *Zeitschrift für Physik B Condensed Matter*, 68(2-3):181–191, 1987.
- [52] Hermann Grabert, Peter Olschowski, and Ulrich Weiss. Quantum decay rates for dissipative systems at finite temperatures. *Phys. Rev. B*, 36(4):1931–1951, 1987.
- [53] Eli Pollak. Sudden theory. *Phys. Rev. B*, 40(4), 1989.
- [54] Ian Affleck. Quantum-Statistical Metastability. *Phys. Rev. Lett.*, 46(6):388–391, 1981.
- [55] Jeremy O. Richardson, Rainer Bauer, and Michael Thoss. Semiclassical Green’s functions and an instanton formulation of electron-transfer rates in the nonadiabatic limit. *J. Chem. Phys.*, 143(13), 2015.
- [56] Jeremy O. Richardson. Derivation of instanton rate theory from first principles. *Arxiv*, 114106(1):1–7, 2015.
- [57] R. P. Feynman and F. L. Vernon. The theory of a general quantum system interacting with a linear dissipative system. *Ann. Phys.*, 24:118, 1963.
- [58] C. Morais Smith and A. O. Caldeira. Generalized Feynman-Vernon approach to dissipative quantum systems. *Physical Review A*, 36(7):3509–3511, 1987.
- [59] Nancy Makri. Effective Non-Oscillatory Propagator For Feynman Path Integration In Real Time. *Chem. Phys. Lett.*, 159(5,6):489–498, 1989.

- [60] Kurt Allinger and Mark A Ratner. Influence functionals: General methodology for subsystem calculations. *Phys. Rev. A*, 39(2):864–880, 1989.
- [61] Jake Iles-Smith, Neill Lambert, and Ahsan Nazir. Environmental dynamics, correlations, and the emergence of noncanonical equilibrium states in open quantum systems. *Phys. Rev. A*, 90:032114, Sep 2014.
- [62] S.N. Shevchenko, S. Ashhab, and Franco Nori. Landau–zener–stuckelberg interferometry. *Physics Reports*, 492(1):1 – 30, 2010.
- [63] David Newman, Florian Mintert, and Ahsan Nazir. Performance of a quantum heat engine at strong reservoir coupling. *Phys. Rev. E*, 95:032139, Mar 2017.
- [64] Hermann Grabert, Peter Schramm, and Gert-Ludwig Ingold. Quantum Brownian motion: The functional integral approach. *Physics Reports*, 168(3):115–207, 1988.
- [65] G W Ford and M Kac. On the Quantum Langevin Equation. *J. Stat. Phys.*, 46:803, 1987.
- [66] C W Gardiner. Quantum noise and quantum Langevin equations. *IBM J. Res. Develop.*, 32:127, 1988.
- [67] K.L. Sebastian. The Classical Description of Scattering From a Quantum System. *Chem. Phys. Lett.*, 81(1):14–16, 1981.
- [68] N G van Kampen. Derivation of the quantum Langevin equation. *J. Molec. Liquids*, 71:97, 1997.
- [69] H. Kleinert. *Path Integrals in Quantum Mechanics , Statistics , Polymer Physics and Financial Markers*. Berlin, 3rd editio edition, 2006.

- [70] H Kleinert and S V Shabanov. Quantum Langevin Equation From Forward-backward Path-integral. *Physics Letters A*, 200(3-4):224–232, 1995.
- [71] K Tsusaka. Generalized quantum Langevin equations from the forward-backward path integral. *Phys. Rev. E*, 59(5 Pt A):4931–8, 1999.
- [72] Chitrak Bhadra and Dhruba Banerjee. System-reservoir theory with anharmonic baths: a perturbative approach. *J. Stat. Mech.*, 2016(4):043404, 2016.
- [73] H. Keith McDowell. Quantum generalized Langevin equation: Explicit inclusion of nonlinear system dynamics. *J. Chem. Phys.*, 112(16):6971, 2000.
- [74] M Carrega, P Solinas, a Braggio, M Sassetti, and U Weiss. Functional integral approach to time-dependent heat exchange in open quantum systems: general method and applications. *New Journal of Physics*, 17(4):045030, 2015.
- [75] Debasish Banerjee, Florian Hebenstreit, Fu-Jiun Jiang, Mark Kon, and Uwe-Jens Wiese. Real-time simulation of dissipation-driven quantum systems. *The 33rd International Symposium on Lattice Field Theory*, 2015.
- [76] Nancy Makri. Exploiting classical decoherence in dissipative quantum dynamics: Memory, phonon emission, and the blip sum. *Chemical Physics Letters*, 593:93–103, 2014.
- [77] Nancy Makri. Dynamics of reduced density matrices: Classical memory versus quantum nonlocality. *J. Chem. Phys.*, 109(8):2994–2998, 1998.
- [78] NIKESH S DATTANI, Felix A Pollock, and David M Wilkins. Analytic influence functionals for numerical Feynman integrals in most open quantum systems. *Quantum Physics Letters*, 1(1):35–45, 2012.

- [79] Scott Habershon, David E Manolopoulos, Thomas E Markland, and Thomas F Miller. Ring-polymer molecular dynamics: quantum effects in chemical dynamics from classical trajectories in an extended phase space. *Annual review of physical chemistry*, 64:387–413, 2013.
- [80] C P Herrero and R Ramírez. Path-integral simulation of solids. *J. Phys. Cond. Mat.*, 26:233201, 2014.
- [81] Jian-Sheng Wang. Quantum thermal transport from classical molecular dynamics. *Phys. Rev. Lett.*, 99(16):160601, 2007.
- [82] Peter P Orth, Adilet Imambekov, and Karyn Le Hur. Nonperturbative stochastic method for driven spin-boson model. *Phys. Rev. B*, 87(014305):119–123, 2013.
- [83] Peter P Orth, Adilet Imambekov, and Karyn Le Hur. Universality in dissipative Landau-Zener transitions. *Phys. Rev. A.*, 82:032118, 2010.
- [84] P Nalbach and M Thorwart. Landau-Zener Transitions in a Dissipative Environment : Numerically Exact Results. *Phys. Rev. Lett.*, 103(November):220401, 2009.
- [85] Jürgen T. Stockburger. Simulating spin-boson dynamics with stochastic Liouville-von Neumann equations. *Chemical Physics*, 296:159–169, 2004.
- [86] L. N. Kantorovich. Generalized langevin equation for solids: I. rigorous derivation and main properties. *Phys. Rev. B*, 78:094304, 2008.
- [87] L. Stella, C. D. Lorenz, and L. Kantorovich. Generalized Langevin equation: An efficient approach to nonequilibrium molecular dynamics of open systems. *Phys. Rev. B*, 89:134303, 2014.

- [88] H. Ness, L. Stella, C. D. Lorenz, and L. Kantorovich. Applications of the generalized Langevin equation: Towards a realistic description of the baths. *Phys. Rev. B*, 91:014301, 2015.
- [89] H. Ness, A. Genina, L. Stella, C. D. Lorenz, and L. Kantorovich. Nonequilibrium processes from generalised Langevin equations: realistic nanoscale systems connected to two thermal baths. *Phys. Rev. B*, 93:174303, 2016.
- [90] L. Kantorovich, H. Ness, L. Stella, and C. D. Lorenz. c-number quantum generalized Langevin equation for an open system. *Phys. Rev. B*, 94(184305), 2016.
- [91] M. Chaichian and A. Demichev. *Path Integrals In Physics*. IoP Publishing, London, 2001.
- [92] J. H. Van Vleck. The correspondence principle in the statistical interpretation of quantum mechanics. *Proceedings of the National Academy of Sciences*, 14(2):178–188, 1928.
- [93] P.A.M. Dirac. The lagrangian in quantum mechanics. *Physikalische Zeitschrift der Sowjetunion*, 3:64–72, 1933.
- [94] M. P. Hobson. *General relativity : an introduction for physicists*. Cambridge University Press, Cambridge, UK New York, 2006.
- [95] Massimo Blasone, Petr Jizba, and Hagen Kleinert. Path-integral approach to 't Hooft's derivation of quantum physics from classical physics. *Physical Review A - Atomic, Molecular, and Optical Physics*, 71(5):1–19, 2005.
- [96] Andreas Wipf. *Statistical approach to quantum field theory : an introduction*. Springer, Berlin, 2013.
- [97] H. F. Trotter. On the product of semi-groups of operators. *Proceedings of the American Mathematical Society*, 10(4):545–545, 1959.

- [98] John R. Klauder. The feynman path integral: An historical slice. In *A Garden of Quanta*, pages 55–76. WORLD SCIENTIFIC, 2003.
- [99] M. Kac. On some connections between probability theory and differential and integral equations. In *Proceedings of the Second Berkeley Symposium on Mathematical Statistics and Probability*, pages 189–215, Berkeley, Calif., 1951. University of California Press.
- [100] I. M. Gel’fand. *Generalized functions*. American Mathematical Society AMS Chelsea Publishing, Providence, Rhode Island, 2016.
- [101] I. M. Gel’fand and A. M. Yaglom. Integration in functional spaces and its applications in quantum physics. *Journal of Mathematical Physics*, 1(1):48–69, 1960.
- [102] R. H. Cameron. A family of integrals serving to connect the wiener and feynman integrals. *Journal of Mathematics and Physics*, 39(1-4):126–140, 1960.
- [103] G. Manfredi and M. R. Feix. Entropy and Wigner functions. *Physical Review E - Statistical Physics, Plasmas, Fluids, and Related Interdisciplinary Topics*, 62(4):4665–4674, 2000.
- [104] Liam Cleary. *a Semiclassical Approach To Quantum Brownian Motion Wigner Phase Space*. PhD thesis, Trinity College Dublin, 2010.
- [105] Denys I. Bondar, Renan Cabrera, and Herschel A. Rabitz. Conceptual inconsistencies in finite-dimensional quantum and classical mechanics. *Physical Review A - Atomic, Molecular, and Optical Physics*, 88(1):1–5, 2013.
- [106] Ingrid Daubechies and John R. Klauder. Quantum-mechanical path integrals with wiener measure for all polynomial hamiltonians. II. *Journal of Mathematical Physics*, 26(9):2239–2256, 1985.

- [107] John R Klauder. Quantization is geometry, after all. *Annals of Physics*, 188(1):120–141, 1988.
- [108] Célia M. A. Dantas, I. A. Pedrosa, and B. Baseia. Harmonic oscillator with time-dependent mass and frequency and a perturbative potential. *Phys. Rev. A*, 45:1320–1324, Feb 1992.
- [109] Swapan Mandal. Classical and quantum harmonic oscillators with time dependent mass and frequency: A new class of exactly solvable model. *Optics Communications*, 386:37–42, mar 2017.
- [110] Nora S. Thornber and Edwin F. Taylor. Propagator for the simple harmonic oscillator. *American Journal of Physics*, 66(11):1022–1024, nov 1998.
- [111] Christopher Bishop. *Pattern recognition and machine learning*. Springer, New York, 2006.
- [112] E. Wigner. On the quantum correction for thermodynamic equilibrium. *Phys. Rev.*, 40:749–759, Jun 1932.
- [113] B O Koopman. Hamiltonian Systems and Transformation in Hilbert Space. *Proceedings of the National Academy of Sciences*, 17(5):315–318, 1931.
- [114] Irán Ramos-Prieto, Alejandro R. Urzúa-Pineda, Francisco Soto-Eguibar, and Héctor M. Moya-Cessa. KvN mechanics approach to the time-dependent frequency harmonic oscillator. *Scientific Reports*, 8(1), may 2018.
- [115] Dariusz Chruściński. Koopman’s approach to dissipation. *Reports on Mathematical Physics*, 57(3):319–332, 2006.
- [116] Steven L Brunton, Bingni W Brunton, Joshua L Proctor, and J Nathan Kutz. Koopman Invariant Subspaces and Finite Linear Representations of Nonlinear Dynamical Systems for Control. *PLOS ONE*, 11(2):1–19, 2016.

- [117] J. v. Neumann. Uber einen satz von herrn m. h. stone. *Ann. Math.*, 33(3):567–573, 1932.
- [118] Denys I. Bondar, Renan Cabrera, Robert R. Lompay, Misha Yu Ivanov, and Herschel A. Rabitz. Operational dynamic modeling transcending quantum and classical mechanics. *Physical Review Letters*, 109(19):1–5, 2012.
- [119] John Hamhalter. *Quantum Measure theory*. Springer, New York, 2003.
- [120] George Baker. Formulation of Quantum Mechanics Based on the Quasi-Probability Distribution Induced on Phase Space. *Physical Review*, 109(6):2198–2206, 1958.
- [121] Thomas L Curtright, David B Fairlie, and Cosmas K Zachos. *a Concise Treatise On Quantum Mechanics In Phase*. World Scientific, 2014.
- [122] H. J. Groenewold. On the principles of elementary quantum mechanics. *Physica*, 12(7):405–460, 1946.
- [123] Renan Cabrera, Denys I. Bondar, Kurt Jacobs, and Herschel A. Rabitz. Efficient method to generate time evolution of the Wigner function for open quantum systems. 042122, 2012.
- [124] Denys I. Bondar, Renan Cabrera, Dmitry V. Zhdanov, and Herschel A. Rabitz. Wigner phase-space distribution as a wave function. *Physical Review A - Atomic, Molecular, and Optical Physics*, 88(5):1–6, 2013.
- [125] Maximilian Schlosshauer. Decoherence, the measurement problem, and interpretations of quantum mechanics. *Rev. Mod. Phys.*, 76:1267–1305, Feb 2005.
- [126] Paul Schilpp. *Albert Einstein: philosopher-scientist*. Open Court, La Salle, Ill, 1970.

- [127] A. Einstein, B. Podolsky, and N. Rosen. Can quantum-mechanical description of physical reality be considered complete? *Phys. Rev.*, 47:777–780, May 1935.
- [128] J. S. Bell. On the einstein podolsky rosen paradox. *Physica*, 1:195–200, Nov 1964.
- [129] Alain Aspect, Philippe Grangier, and Gérard Roger. Experimental tests of realistic local theories via bell’s theorem. *Phys. Rev. Lett.*, 47:460–463, Aug 1981.
- [130] Gregor Weihs, Thomas Jennewein, Christoph Simon, Harald Weinfurter, and Anton Zeilinger. Violation of bell’s inequality under strict einstein locality conditions. *Phys. Rev. Lett.*, 81:5039–5043, Dec 1998.
- [131] M. A. Rowe, D. Kielpinski, V. Meyer, C. A. Sackett, W. M. Itano, C. Monroe, and D. J. Wineland. Experimental violation of a Bell’s inequality with efficient detection. *Nature*, 409:791–794, February 2001.
- [132] S. Gröblacher, T. Paterek, R. Kaltenbaek, Č. Brukner, M. Żukowski, M. Aspelmeyer, and A. Zeilinger. An experimental test of non-local realism. *Nature*, 446:871–875, April 2007.
- [133] M. Giustina, A. Mech, S. Ramelow, B. Wittmann, J. Kofler, J. Beyer, A. Lita, B. Calkins, T. Gerrits, S. W. Nam, R. Ursin, and A. Zeilinger. Bell violation using entangled photons without the fair-sampling assumption. *Nature*, 497:227–230, May 2013.
- [134] Lynden K. Shalm, Evan Meyer-Scott, Bradley G. Christensen, Peter Bierhorst, Michael A. Wayne, Martin J. Stevens, Thomas Gerrits, Scott Glancy, Deny R. Hamel, Michael S. Allman, Kevin J. Coakley, Shellee D. Dyer, Carson Hodge, Adriana E. Lita, Varun B. Verma, Camilla Lambrocco, Edward

- Tortorici, Alan L. Migdall, Yanbao Zhang, Daniel R. Kumor, William H. Farr, Francesco Marsili, Matthew D. Shaw, Jeffrey A. Stern, Carlos Abellán, Waldimar Amaya, Valerio Pruneri, Thomas Jennewein, Morgan W. Mitchell, Paul G. Kwiat, Joshua C. Bienfang, Richard P. Mirin, Emanuel Knill, and Sae Woo Nam. Strong loophole-free test of local realism. *Phys. Rev. Lett.*, 115:250402, Dec 2015.
- [135] BIG Bell Test Collaboration. Challenging local realism with human choices. *Nature*, 557:212–216, May 2018.
- [136] Robert W. Spekkens. Evidence for the epistemic view of quantum states: A toy theory. *Phys. Rev. A*, 75:032110, Mar 2007.
- [137] G. M. G. McCaul, C. D. Lorenz, and L. Kantorovich. Partition-free approach to open quantum systems in harmonic environments: An exact stochastic liouville equation. *Phys. Rev. B*, 95:125124, Mar 2017.
- [138] W. K. Wootters and W. H. Zurek. A single quantum cannot be cloned. *Nature*, 299(5886):802–803, oct 1982.
- [139] Stefanie Hilt, Benedikt Thomas, and Eric Lutz. Hamiltonian of mean force for damped quantum systems. *Phys. Rev. E*, 84:031110, Sep 2011.
- [140] Udo Seifert. First and second law of thermodynamics at strong coupling. *Phys. Rev. Lett.*, 116:020601, Jan 2016.
- [141] L. Isserlies. On a formula for the product-moment coefficient of any order of a normal frequency distribution in any number of variables. *Biometrika*, 12(1-2):134–139, nov 1918.

- [142] Jeremy M. Moix, Yang Zhao, and Jianshu Cao. Equilibrium-reduced density matrix formulation: Influence of noise, disorder, and temperature on localization in excitonic systems. *Phys. Rev. B*, 85(11):1–14, 2012.
- [143] Yoshitaka Tanimura. Reduced hierarchical equations of motion in real and imaginary time: Correlated initial states and thermodynamic quantities. *J. Chem. Phys.*, 141(4), 2014.
- [144] G. M. G. McCaul, C. D. Lorenz, and L. Kantorovich. Driving spin-boson models from equilibrium using exact quantum dynamics. *Phys. Rev. B*, 97:224310, Jun 2018.
- [145] Julian Schmidt, Alex Meistrenko, Hendrik van Hees, Zhe Xu, and Carsten Greiner. Simulation of stationary gaussian noise with regard to the langevin equation with memory effect. *Phys. Rev. E*, 91:032125, Mar 2015.
- [146] A.V Oppenheim and R.w Schafer. *Discrete-Time Signal Processing*. Pearson, Harlow, 2014.
- [147] Matteo Carrega, Paolo Solinas, Maura Sassetti, and Ulrich Weiss. Energy exchange in driven open quantum systems at strong coupling. *Phys. Rev. Lett.*, 116:240403, Jun 2016.
- [148] Peter P. Orth, David Roosen, Walter Hofstetter, and Karyn Le Hur. Dynamics, synchronization, and quantum phase transitions of two dissipative spins. *Phys. Rev. B*, 82:144423, Oct 2010.
- [149] David R Reichman, Frank L H Brown, and Peter Neu. Cumulant expansions and the spin-boson problem. *Phys. Rev. E*, 55(3):2328–2337, 1997.
- [150] Michael L Wall, Arghavan Safavi-naini, and Ana Maria Rey. Simulating generic spin-boson models with matrix product states. *Phys. Rev. A*, 94:053637, 2016.

- [151] Yosuke Kayanuma and Hiroyuki Nakayama. Nonadiabatic transition at a level crossing with dissipation. *Physical Review B*, 57(20):13099–13112, 1998.
- [152] D. Dylewsky, J. K. Freericks, M. L. Wall, A. M. Rey, and M. Foss-Feig. Non-perturbative calculation of phonon effects on spin squeezing. *Physical Review A*, 93(1):1–12, 2016.
- [153] David P. DiVincenzo and Daniel Loss. Rigorous Born approximation and beyond for the spin-boson model. *Physical Review B - Condensed Matter and Materials Physics*, 71(3):1–10, 2005.
- [154] Clarence Zener. Non-Adiabatic Crossing of Energy Levels. *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, 137(833):696–702, 1932.
- [155] Curt Wittig. The Landau-Zener Formula. *J. Phys. Chem. B*, 109(17):8428, 2005.
- [156] Alberto G Rojo. Matrix exponential solution of the Landau-Zener problem. *Arxiv*, page 1004.2914, 2010.
- [157] A. Bambini and P. R. Berman. Analytic solutions to the two-state problem for a class of coupling potentials. *Physical Review A*, 23(5):2496–2501, 1981.
- [158] C. E. Carroll and F. T. Hioe. Analytic solution of the two-state problem. *Physical Review A*, 41(5):2835–2836, 1990.
- [159] Vyacheslav V Stepanov, Gerhard Müller, and Joachim Stolze. Quantum integrability and nonintegrability in the spin-boson model. *Phys. Rev. E*, 77:066202, 2008.

- [160] F Guinea, V Hakim, and A Muramatsu. Diffusion and Localization of a Particle in a Periodic Potential Coupled to a Dissipative Environment. *Phys. Rev. Lett.*, 54(4):263–266, 1985.
- [161] F. Lesage and H. Saleur. Boundary interaction changing operators and dynamical correlations in quantum impurity problems. *Phys. Rev. Lett.*, 80:4370–4373, May 1998.
- [162] Reinhold Egger, Hermann Grabert, and Ulrich Weiss. Crossover from coherent to incoherent dynamics in damped quantum systems. *Phys. Rev. E*, 55:R3809–R3812, Apr 1997.
- [163] Matthias Vojta. Impurity quantum phase transitions Impurity quantum phase transitions. *Philosophical Magazine*, 86(13):1807–1846, 2006.
- [164] M. Blume, V. J. Emery, and A Luther. Spin-Boson Systems: One-Dimensional Equivalents and the Kondo Problem. *Phys. Rev. Lett.*, 25(7):4–7, 1970.
- [165] Jie Liu, Libin Fu, Bi-yiao Ou, Shi-gang Chen, Dae-il Choi, Biao Wu, and Qian Niu. Theory of nonlinear Landau-Zener tunneling. *Phys. Rev. A.*, 66:023404, 2002.
- [166] Yuriy Makhlin, Gerd Schön, and Alexander Shnirman. Quantum-state engineering with josephson-junction devices. *Rev. Mod. Phys.*, 73:357–400, May 2001.
- [167] Davide Valenti, Claudio Guarcello, and Bernardo Spagnolo. Switching times in long-overlap Josephson junctions subject to thermal fluctuations and non-Gaussian noise sources. *Physical Review B - Condensed Matter and Materials Physics*, 89(21):1–15, 2014.

- [168] Peter P. Orth, Ivan Stanic, and Karyn Le Hur. Dissipative quantum ising model in a cold-atom spin-boson mixture. *Phys. Rev. A*, 77:051601, May 2008.
- [169] Anupam Garg, José Nelson Onuchic, and Vinay Ambegaokar. Effect of friction on electron transfer in biomolecules. *The Journal of Chemical Physics*, 83(9):4491–4503, 1985.
- [170] K. Le Hur. Entanglement entropy, decoherence, and quantum phase transitions of a dissipative two-level system. *Annals of Physics*, 323(9):2208 – 2240, 2008.
- [171] Luca Magazzu, Angelo Carollo, Bernardo Spagnolo, and Davide Valenti. Quantum dissipative dynamics of a bistable system in the sub-ohmic to super-ohmic regime. *JSM: Theory and Experiment*, 2016(5):054016, 2016.
- [172] Jan R. Rubbmark, Michael M. Kash, Michael G. Littman, and Daniel Kleppner. Dynamical effects at avoided level crossings: A study of the landau-zener effect using rydberg atoms. *Phys. Rev. A*, 23:3107–3117, Jun 1981.
- [173] Abraham J. Olson, Su-Ju Wang, Robert J. Niffenegger, Chuan-Hsun Li, Chris H. Greene, and Yong P. Chen. Tunable landau-zener transitions in a spin-orbit-coupled bose-einstein condensate. *Phys. Rev. A*, 90:013616, Jul 2014.
- [174] Martijn Wubs, Keiji Saito, Sigmund Kohler, Peter Hänggi, and Yosuke Kayanuma. Gauging a quantum heat bath with dissipative landau-zener transitions. *Phys. Rev. Lett.*, 97:200404, Nov 2006.
- [175] Keiji Saito, Martijn Wubs, Sigmund Kohler, Yosuke Kayanuma, and Peter Hänggi. Dissipative landau-zener transitions of a qubit: Bath-specific and universal behavior. *Phys. Rev. B*, 75:214308, Jun 2007.
- [176] Hassan Shapourian. Dynamical renormalization-group approach to the spin-boson model. *Phys. Rev. A*, 93:032119, Mar 2016.

- [177] R. Mankin, K. Laas, and A. Sauga. Generalized langevin equation with multiplicative noise: Temporal behavior of the autocorrelation functions. *Phys. Rev. E*, 83:061131, Jun 2011.
- [178] H. Nakajima and S. Furui. A new algorithm for numerical simulation of langevin equations. *Nuclear Physics B - Proceedings Supplements*, 53(1):983 – 986, 1997. Lattice 96.
- [179] Kun Lü and Jing-Dong Bao. Numerical simulation of generalized langevin equation with arbitrary correlated noise. *Phys. Rev. E*, 72:067701, Dec 2005.
- [180] H. J.D. Miller and J. Anders. Entropy production and time asymmetry in the presence of strong interactions. *Physical Review E*, 95(6):1–7, 2017.
- [181] Harry J.D. Miller and Janet Anders. Time-reversal symmetric work distributions for closed quantum dynamics in the histories framework. *New Journal of Physics*, 19(6), 2017.
- [182] Ian J Ford. Measures of thermodynamic irreversibility in deterministic and stochastic dynamics. *New J. Phys.*, 17(7):075017, 2015.
- [183] J M R Parrondo, C Van den Broeck, and R Kawai. Entropy production and the arrow of time. *New Journal of Physics*, 11(7):73008, 2009.
- [184] H. R. Brown, W. Myrvold, and J. Uffink. Boltzmann’s H-theorem, its discontents, and the birth of statistical mechanics. *Studies in the History and Philosophy of Modern Physics*, 40:174–191, 2009.
- [185] T. B. Batalhão, A. M. Souza, R. S. Sarthour, I. S. Oliveira, M. Paternostro, E. Lutz, and R. M. Serra. Irreversibility and the Arrow of Time in a Quenched Quantum System. *Physical Review Letters*, 115(19):1–5, 2015.

- [186] Elliott H. Lieb and Jakob Yngvason. A fresh look at entropy and the second law of thermodynamics. *Physics Today*, 53(4):32–37, apr 2000.
- [187] ET Edwin T. Jaynes. The Gibbs paradox. In *Maximum entropy and bayesian methods*, volume 17, pages 1–21. Springer, 1992.
- [188] Elliott H Lieb and Jakob Yngvason. The physics and mathematics of the second law of thermodynamics. *Physics Reports*, 310(1):1–96, 1999.
- [189] L.D Landau and E.M. Lifshitz. *Course of Theoretical Physics: Statistical Physics*, volume 5. Elsevier, London, 1951.
- [190] Guy Bonneau, Jacques Faraut, and Galliano Valent. Self-adjoint extensions of operators and the teaching of quantum mechanics. *American Journal of Physics*, 69:322, 2001.
- [191] Xie Chen, Yuan Ming Lu, and Ashvin Vishwanath. Symmetry-protected topological phases from decorated domain walls. *Nature Communications*, 5:1–11, 2014.
- [192] Xie Chen, Zheng Cheng Gu, Zheng Xin Liu, and Xiao Gang Wen. Symmetry-protected topological orders in interacting bosonic systems. *Science*, 338(6114):1604–1606, 2012.
- [193] Mostafa Tanhayi Ahari, Gerardo Ortiz, and Babak Seradjeh. On the Role of Self-Adjointness in the Continuum Formulation of Topological Quantum Phases. *American Journal of Physics*, 84:858, 2016.
- [194] Vanilse S. Araujo, F. A. B. Coutinho, and J. Fernando Perez. Operator domains and self-adjoint operators. *American Journal of Physics*, 72(2):203–213, 2004.
- [195] David H. Berman. Boundary effects in quantum physics. *American Journal of Physics*, 59:937, 1991.

- [196] Anton Z. Capri. Self adjointness and spontaneously broken symmetry. *American Journal of Physics*, 45(9):823–825, 1977.
- [197] Carl M. Bender, Dorje C. Brody, and Markus P. Müller. Hamiltonian for the Zeros of the Riemann Zeta Function. *Physical Review Letters*, 118(13):1–5, 2017.
- [198] Michael Reed and Barry Simon. *Methods of Modern Mathematical Physics*, volume 1. Elsevier, London, 1980.
- [199] Michael Reed and Barry Simon. *Methods of Modern Mathematical Physics*, volume 2. Elsevier, London, 1980.
- [200] Joseph Samuel and Rajendra Bhandari. General Setting for Berry’s phase. *Phys. Rev. Lett.*, 60(23):2339, 1988.
- [201] J H HANNAY. Angle Variable Holonomy in Adiabatic Excursion of an Integrable Hamiltonian. *J. Phys. A*, 18(2):221–230, 1985.
- [202] O V Usatenko, J-P Provost, and G Vallée. A comparative study of the Hannay’s angles associated with a damped harmonic oscillator and a generalized harmonic oscillator. *Journal of Physics A: Mathematical and General*, 29(10):2607, 1996.
- [203] M. Revenga, I. Zuniga, and P. Espanol. Boundary conditions in dissipative particle dynamics. *Computer Physics Communications*, 121-122:309 – 311, 1999.
- [204] D. Grassano, O. Pulci, V. O. Shubnyi, S. G. Sharapov, V. P. Gusynin, A. V. Kavokin, and A. A. Varlamov. Detection of topological phase transitions through entropy measurements: The case of germanene. *Phys. Rev. B*, 97:205442, May 2018.
- [205] Sebastian D. Huber. Topological mechanics. *Nature Physics*, 12(7):621–623, 2016.

- [206] Emil Prodan and Camelia Prodan. Topological phonon modes and their role in dynamic instability of microtubules. *Physical Review Letters*, 103(24):1–4, 2009.
- [207] Nina Berg, Kira Joel, Miriam Koolyk, and Emil Prodan. Topological phonon modes in filamentary structures. *Physical Review E - Statistical, Nonlinear, and Soft Matter Physics*, 83(2):1–8, 2011.
- [208] ET Edwin T. Jaynes. Gibbs vs Boltzmann Entropies. *American Journal of Physics*, 33(5):391–398, 1965.
- [209] Nancy Makri. The Linear Response Approximation and Its Lowest Order Corrections An Influence Functional Approach. *J. Phys. Chem. B*, 103(15):2823–2829, 1999.
- [210] Peter L. Walters and Nancy Makri. Quantum Classical Path Integral Simulation of Ferrocene-Ferrocenium Charge Transfer in Liquid Hexane. *Physical Chemistry Letters*, 6(24):4959–4965, 2015.
- [211] Hermann Grabert and Michael Thorwart. Quantum mechanical response to a driven caldeira-leggett bath. *Phys. Rev. E*, 98:012122, Jul 2018.
- [212] Robert J. Rubin. Momentum autocorrelation functions and energy transport in harmonic crystals containing isotopic defects. *Phys. Rev.*, 131:964–989, Aug 1963.
- [213] Luca Magazzù, Davide Valenti, Angelo Carollo, and Bernardo Spagnolo. Multi-state quantum dissipative dynamics in sub-ohmic environment: The strong coupling regime. *Entropy*, 17(4):2341–2354, 2015.
- [214] Luca Magazzù, Davide Valenti, Bernardo Spagnolo, and Milena Grifoni. Dissipative dynamics in a quantum bistable system: Crossover from weak to strong

- damping. *Physical Review E - Statistical, Nonlinear, and Soft Matter Physics*, 92(3), 2015.
- [215] D. Valenti, L. Magazzù, P. Caldara, and B. Spagnolo. Stabilization of quantum metastable states by dissipation. *Physical Review B - Condensed Matter and Materials Physics*, 91(23):1–7, 2015.
- [216] Luca Magazzù, Peter Hänggi, Bernardo Spagnolo, and Davide Valenti. Quantum resonant activation. *Physical Review E*, 95(4):1–13, 2017.
- [217] Max Born. The statistical interpretation of quantum mechanics. In *Nobel Lectures in Physics 1942 – 1962*. WORLD SCIENTIFIC, 1998.
- [218] Eugene P. Wigner. The unreasonable effectiveness of mathematics in the natural sciences. richard courant lecture in mathematical sciences delivered at new york university, may 11, 1959. *Communications on Pure and Applied Mathematics*, 13(1):1–14.
- [219] P W Anderson. More Is Different. *Science*, 177(4047):393–396, 1972.